

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

09/890711

INTERNATIONAL APPLICATION NO.
PCT/JP00/08298INTERNATIONAL FILING DATE
24 November 2000PRIORITY CLAIMED
12 February 1999TITLE OF INVENTION
STYRYL DYESAPPLICANT(S) FOR DO/EO/US
Chiaki KASADA et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information.

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ The US has been elected in a Demand by the expiration of 19 months from the priority date (PCT Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☐ is attached hereto (required only if not transmitted by the International Bureau).
 - b. ☒ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☒ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An Assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information.
 - ☒ Courtesy copy of the first page of the International Publication (WO 01/40382).
 - ☒ Formal drawings, 2 sheets, Figures 1-3.
 - ☒ Courtesy Copy of the International Search Report.
 - ☒ The application is (or will be) assigned to: KABUSHIKI KAISHA HAYASHIBARA SEIBUTSU KAGAKU KENKYUJO, whose address is 2-3, Shimoishu 1-chome, Okayama-shi, Okayama, Japan.

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

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International Application No.

PCT/JP00/08298

Attorney's Docket No.

KASADA=4

17. [xx] The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a)(1)-(5):

Neither international preliminary examination fee (37 CFR 1.482)
nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO
and International Search Report not prepared by the EPO or JPO. **\$1000.00**

International preliminary examination fee (37 CFR 1.482) not paid to
USPTO but International Search Report prepared by the EPO or JPO. **\$860.00**

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but
international search fee (37 CFR 1.445(a)(2)) paid to USPTO. **\$710.00**

International preliminary examination fee paid to USPTO (37 CFR 1.482)
but all claims did not satisfy provisions of PCT Article 33(1)-(4). **\$690.00**

International preliminary examination fee paid to USPTO (37 CFR 1.482)
and all claims satisfied provisions of PCT Article 33(1)-(4). **\$100.00**

CALCULATIONS PTO USE ONLY**ENTER APPROPRIATE BASIC FEE AMOUNT =**

\$ 860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than [] 20 [] 30
months from the earliest claimed priority date (37 CFR 1.492(e)).

Claims as Originally Presented	Number Filed	Number Extra	Rate
Total Claims	14 - 20		X \$18.00
Independent Claims	1 - 3		X \$80.00
Multiple Dependent Claims (if applicable)			+ \$270.00

TOTAL OF ABOVE CALCULATIONS =

\$ 860.00

Claims After Post Filing Prel. Amend	Number Filed	Number Extra	Rate
Total Claims	20 - 20		X \$18.00
Independent Claims	1 - 3		X \$78.00

TOTAL OF ABOVE CALCULATIONS =

\$ 860.00

Reduction of 1/3 for filing by small entity, if applicable. Applicant claims small entity
status. See 37 CFR 1.27

SUBTOTAL =

\$ 860.00

Processing fee of \$130.00 for furnishing the English translation later than [] 20 [] 30
months from the earliest claimed priority date (37 CFR 1.492(f)).

TOTAL NATIONAL FEE =

\$ 860.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). **\$40.00** per property +

TOTAL FEES ENCLOSED =

\$ 860.00

Amount to be:
refunded \$
charged \$

- a. [] A check in the amount of \$ _____ to cover the above fees is enclosed.
b. [X] Credit Card Payment Form (PTO-2038), authorizing payment in the amount of \$ 860.00, is attached.
c. [] Please charge my Deposit Account No. **02-4035** in the amount of \$ _____ to cover the above fees.
A duplicate copy of this sheet is enclosed.
d. [XX] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment
to Deposit Account No. **02-4035**. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or
(b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO

BROWDY AND NEIMARK, P.L.L.C.
624 NINTH STREET, N.W., SUITE 300
WASHINGTON, D.C. 20001
TEL: (202) 628-5197
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Date of this submission: August 2, 2001

Form PTO-1390 (as slightly revised by Browdy and Neimark)



SIGNATURE

Anne M. Kornbau

NAME

25.884

REGISTRATION NUMBER

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

ATTY.'S DOCKET: KASADA=4

In re Application of:)	Art Unit:
Chiaki KASADA et al.)	
)	Examiner:
)	
I.A. No.: PCT/JP00/08298)	Washington, D.C.
)	
Filed: November 24, 2000)	August 2, 2001
)	
For: STYRYL DYES)	

SUPPLEMENTAL PRELIMINARY AMENDMENT

Honorable Commissioner for Patents
Washington, D.C. 20231

Concurrent with filing of the above-identified
application, please enter the following Preliminary Amendment:

IN THE CLAIMS

Please insert new claims 15-20 as follows:

15. (New) The light absorbent of claim 7 which
further contains one or more appropriate light-resistant
improvers.

16. (New) The light absorbent of claim 7 which is
sensitive to a visible light with a wavelength of 450 nm or
less when formed in a thin layer.

17. (New) The light absorbent of claim 8 which is
sensitive to a visible light with a wavelength of 450 nm or
less when formed in a thin layer.

18. (New) The optical recording medium of claim 11
which further contains one or more appropriate light-resistant

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improvers in a recording layer.

19. (New) The optical recording medium of claim 11 which uses a laser beam with a wavelength of 450 nm or less as a writing light.

20. (New) The optical recording medium of claim 12 which uses a laser beam with a wavelength of 450 nm or less as a writing light.

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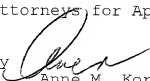
REMARKS

Applicant submits that the invention is new and unobvious and not disclosed by the cited art. Accordingly, Applicant respectfully solicits the Examiner=s early review and issuance of this application.

Respectfully submitted,

BROWDY AND NEIMARK, P.L.L.C.
Attorneys for Applicant(s)

By



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AMK:nmp

F:\A\sausa\kasada4\ptol\2nd prelim amends

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

ATTY.'S DOCKET: KASADA=4

In re Application of:)	Art Unit:
Chiaki KASADA et al.)	Examiner:
)	
I.A. No.: PCT/JP00/08298)	Washington, D.C.
)	
Filed: November 24, 2000)	August 2, 2001
)	
For: STYRYL DYES)	

PRELIMINARY AMENDMENT

Honorable Commissioner for Patents
Washington, D.C. 20231

Concurrent with filing of the above-identified
application, please enter the following Preliminary Amendment:

IN THE SPECIFICATION

After the title please insert the following
paragraph:

REFERENCE TO RELATED APPLICATIONS

The present application is the national stage under
35 U.S.C. 371 of international application PCT/JP00/08298,
filed November 24, 2000 which designated the United States,
and which international application was not published under
PCT Article 21(2) in the English language.

Page 1, before the title, please delete DESCRIPTION.

Page 1, after the title, please delete Technical
Field, and replace it with Field of the Invention.

Page 1, before paragraph 2, delete Background Art,

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and replace it with Background of the Invention.

Page 1, please amend paragraph 2 as follows:

In a multimedia age, optical recording media such as CD-R (a write-once memory using compact disc) and DVD-R (a write-once memory using digital video disc) are now of great importance. Most of the conventional optical recording media can be roughly classified into inorganic optical recording media which have recording layers composed of inorganic substances such as tellurium, selenium, rhodium, carbon, or carbon sulfide; and organic optical recording media which have recording layers mainly composed of light absorbents containing organic dye compounds.

Page 1, please amend paragraph 3 as follows:

Among these optical recording media, organic optical recording media can be usually prepared by dissolving a polymethine dye in an organic solvent such as 2,2,3,3-tetrafluoro-1-propanol (abbreviated as "TFP" hereinafter), spin coating the solution onto the surface of a polycarbonate substrate, drying the solution to form a recording layer, and sequentially forming and coating onto the surface of the recording layer a reflection layer comprising a metal such as gold, silver or copper, and a protection layer comprising an ultraviolet ray hardening resin. When compared with inorganic

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optical recording media, organic ones may have the drawback that their recording layers are susceptible to change under environmental lights such as reading and natural light. Organic optical recording media, however, have the advantage that they can be made into optical recording media at a lesser cost because their recording layers can be directly formed by coating light absorbents in solution on the surface of substrates. Further, organic optical recording media composed of organic materials are now mainly used as low-cost optical recording media because they are substantially free of corrosion even when contacted by moisture or sea water and because information recorded therein in a prescribed format can be read out by using commercially available read-only readers after establishing thermal-deformation-type optical recording media, a kind of organic optical recording media.

Page 2, please amend paragraph 1 as follows:

What is urgently required in organic optical recording media is to increase their storage capacity to suit to this multimedia age. The research for such an increment, which is now being eagerly continued in this field, is to increase the recording capacity per one side from 4.7 GB (giga bite) to 15 GB or more by shortening the wavelength for writing information to 450 nm or less from the wavelengths of 635-650 nm which are commonly used. The high-density optical recording media have the

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capacity to record movie and animations for six hours in picture quality of standard television or for two hours even in relatively-high picture quality of high definition television. However, since most organic dye compounds now used in the conventional optical recording media cannot be used with visible light with a wavelength of 450 nm or less, they could not fulfil the need for high-storage density required in many fields.

Page 3, before paragraph 2, delete Summary of Invention and replace it with Summary of the Invention.

Page 3, please amend paragraph 2 as follows:

The present inventors eagerly studied and screened compounds. As a result, they found that specific styryl dyes (may be called "styryl dyes" hereinafter), which are obtainable through a step of reacting a quaternary ammonium salt of nitrogen heterocyclic compound having an active methyl- or active methylene- group with an aldehyde compound, have absorption maxima at a wavelength of 400 nm or less and substantially absorb visible light with a wavelength around 400 nm. The present inventors confirmed that the styryl dyes, which substantially absorb visible light with a wavelength of 450 nm or less when formed in a thin layer, form remarkably minute pits at a relatively high density in optical recording media by irradiating with a laser beam with a wavelength of 450 nm or less. The

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present invention was made based on the creation of novel styryl dyes and the discovery of their industrially useful characteristics.

Page 4, before paragraph 1, delete Brief Description of Drawings and replace it with Brief Description of the Drawings.

Page 4, please amend paragraph 1 as follows:

FIG. 1 is the visible absorption spectra of one of the styryl dyes of the present invention when in a solution form and in a thin layer form, respectively.

Page 4, please amend paragraph 2 as follows:

FIG. 2 is the visible absorption spectra of another styryl dye of the present invention when in a solution form and in a thin layer form, respectively.

Page 4, please amend paragraph 3 as follows:

FIG. 3 is the visible absorption spectra of a conventional related compound when in a solution form and in a thin layer form, respectively.

Page 4, before paragraph 4, delete Best Mode for carrying out the Invention and replace it with Detailed Description of the Invention.

Page 4, please amend paragraph 5 as follows:

In Formula 1, R_1 represents a hydrogen atom, an aliphatic hydrocarbon group, ether group, acyl group, halogen, or cyano group. The aliphatic hydrocarbon group in R_1 represented that having up to six carbon atoms, usually, from one to five carbon atoms such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, *sec*-butyl, *tert*-butyl, pentyl, isopentyl, neopentyl, or *tert*-pentyl group. Examples of the ether groups are methoxy, ethoxy, propoxy, isopropoxy, butoxy, *tert*-butoxy, pentoxy, and phenoxy groups. Examples of the acyl groups are formyl, acetyl, propionyl, butyryl, *tert*-butyryl, benzoyl, *o*-tolyl, *m*-tolyl, and *p*-tolyl groups. Examples of the halogens are fluorine, chlorine, bromine, and iodine. One or more of the hydrogens in the aliphatic hydrocarbon group, ether group, and acyl group may be replaced with a short-chain aliphatic hydrocarbon group such as methyl, ethyl, or propyl group; a short-chain ether group such as methoxy, ethoxy, or propoxy group; or a halogen such as fluorine, chlorine, bromine, or iodine.

Page 6, please amend paragraph 1 as follows:

Throughout Formulae 2 to 7, A represents a monocyclic or polycyclic-aromatic-ring or heterocycle. Examples of the aromatic ring are benzene and naphthalene rings. Examples of the heterocycle are pyridine, quinoline, and quinoxaline rings. The

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aromatic ring and heterocycle may have one or more substituents, for example, a halogen atom such as fluorine, chlorine, bromine, or iodine atom; a short-chain aliphatic hydrocarbon group such as methyl, trifluoromethyl, ethyl, propyl, isopropyl, butyl, isobutyl, sec-butyl, or tert-butyl group; a short-chain ether group such as methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, butoxy, or tert-butoxy group; an alkylaminosulfonyl group such as methylaminosulfonyl, dimethylaminosulfonyl, ethylaminosulfonyl, diethylaminosulfonyl, propylaminosulfonyl, dipropylaminosulfonyl, butylaminosulfonyl, or dibutylaminosulfonyl group; an aromatic hydrocarbon group such as phenyl, biphenyl, *o*-tolyl, *m*-tolyl, *p*-tolyl, xylyl, mesityl, *o*-cumenyl, *m*-cumenyl, *p*-cumenyl, or naphthyl group; an ester group such as methoxycarbonyl, trifluoromethoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, acetoxyl, trifluoroacetoxyl, or benzoyloxy group; nitro group, cyano group, carboxy group, or sulfo group. When A is not present in Formulae 2 to 7, one or more substituents similar to those that are bound to A may be in the position where A is located.

Page 7, please amend paragraph 1 as follows:

Throughout Formulae 2 to 8, R₂ represents an aliphatic hydrocarbon group selected from those having up to eight carbon atoms, usually, up to six carbon atoms such as methyl, ethyl, propyl, isopropyl, isopropenyl, 1-propenyl, 2-propenyl, 2-

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propynyl, butyl, isobutyl, *sec*-butyl, *tert*-butyl, 2-butenyl, 2-butenyl, 1,3-butadienyl, pentyl, isopentyl, neopentyl, 1-methylpentyl, 2-methylpentyl, 2-pentenyl, 2-pentynyl, 2-penten-4-ynyl, hexyl, or isohexyl group. The aliphatic hydrocarbon group may have one or more substituents, for example, a halogen such as fluorine, chlorine, bromine, or iodine; an ether group such as methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, butoxy, *tert*-butoxy, benzyloxy, or phenoxy group; an ester group such as methoxycarbonyl, trifluoromethoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, acetoxyl, trifluoroacetoxyl, or benzoyloxy group; a carboxy group; a sulfo group; a hydroxy group; an amido group such as acetylamino or benzoylamino group; an aromatic hydrocarbon group such as phenyl, biphenyl, *o*-tolyl, *m*-tolyl, *p*-tolyl, xylyl, mesityl, *o*-cumenyl, *m*-cumenyl, *p*-cumenyl, or naphthyl group; or heterocyclic group such as 2-pyridyl, 2-quinolyl, 2-tetrahydropyranyl, 2,2-dimethyl-1,3-dioxolane-4-yl, 1,3-dioxolane-2-yl, 3,5-dimethylisooxazole-4-yl, 3-piperidinyl, piperidino, morpholino, 1-piperazinyl, pyrrolidine-1-yl, 1-methyl-2-pyrrolidinyl, 2-benzoimidazolyl, 5-uracil, or benzotriazole-1-yl group.

Page 8, please amend paragraph 1 as follows:

In Formula 4, R_3 represents a hydrogen atom or an aliphatic hydrocarbon group which is identical to or different from R_2 . Representative examples of the aliphatic hydrocarbon

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group are short-chain groups such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, *sec*-butyl, or *tert*-butyl group, which may have one or more substituents, for example, an ether group such as methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, butoxy, or *tert*-butoxy group; an ester group such as methoxycarbonyl, trifluoromethoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, acetoxy, trifluoroacetoxy, or benzoyloxy group; or an aromatic hydrocarbon group such as phenyl, *o*-tolyl, *m*-tolyl, *p*-tolyl, xyl, mesityl, *o*-cumenyl, *m*-cumenyl, *p*-cumenyl, or nitrophenyl group.

Page 8, please amend paragraph 2 as follows:

Furthermore, in Formula 1, f_2 represents a monocyclic-or polycyclic-aromatic ring or heterocycle which may include one or more nitrogen atoms into the ring, for example, benzene, pyridine, pyrimidine, pyridazine, naphthalene, quinoline, or quinoxaline ring. The aromatic ring and heterocycle may have one or more substituents. The substituents are selected from those having up to eight carbon atoms, usually, one to six carbon atoms, for example, an aliphatic hydrocarbon group such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, *sec*-butyl, *tert*-butyl, pentyl, isopentyl, neopentyl, *tert*-pentyl, 1-methylpentyl, 2-methylpentyl, hexyl, or isohexyl group; a short-chain ether group such as methoxy, trifluoromethoxy, ethoxy,

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propoxy, isopropoxy, buthoxy, or *tert*-buthoxy group; an amido group such as acetylamino or benzoylamino group; a halogen such as fluorine, chlorine, bromine, or iodine; and cyano group, nitro group, hydroxy group, sulfo group, or carboxy group. When ϕ_2 is a nitrogen-atom-containing heterocycle, the nitrogen atom may combine with the similar aliphatic hydrocarbon group as in R_2 in ϕ_1 to form an ammonium salt. When ϕ_2 has two hydroxy groups as a substituent and the hydroxy groups combine with adjacent carbon atoms in ϕ_2 , the hydroxy groups may react with an oxo compound such as formalin to form a cyclic structure such as a dioxole ring.

Page 9, please amend paragraph 1 as follows:

X^- in Formula 1 represents an arbitrary counter ion and is usually selected from inorganic acid ions such as fluoride, chloride, bromide, iodide, perchloric acid, periodic acid, phosphoric acid hexafluoride, antimony hexafluoride, tin acid hexafluoride, phosphoric acid, fluoroboric acid, and tetrafluoroboric acid ions; organic acid ions such as thiocyanic acid, benzensulfonic acid, naphthalenesulfonic acid, naphthalenedisulfonic acid, *p*-toluenesulfonic acid, alkylsulfonic acid, benzenecarboxylic acid, alkylcarboxylic acid, trihaloalkylcarboxylic acid, alkylsulfonic acid, trihaloalkylsulfonic acid, nicotinic acid, and

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tetracyanoquinonedimethane ions; and organic metal complex ions such as azo, bisphenyldithiol, thiocatechol chelate, thiobisphenolate chelate, and bisdiol- α -diketone. "n" is the number of X⁻ to balance the electric charge in the styryl dyes, and is usually an integer selected from one and two.

Page 10, please amend paragraph 1 as follows:

The present invention relates to styryl dyes having the structures as mentioned above, particularly, to those which have absorption maxima at a wavelength of 400 nm or less, and desirably those which substantially absorb a visible light with a wavelength of 450 nm or less when formed in a thin layer. Concrete examples of the styryl dyes are those represented by Chemical Formulae 1 to 60. Since all of them have absorption maxima at a wavelength of 400 nm or less and substantially absorb a visible light with a wavelength around 400 nm, they have diversified uses in the fields which need organic compounds that absorb the aforesaid visible light. Among the styryl dyes, those, which substantially absorb a visible light with a wavelength of 450 nm or less when formed in a thin layer, particularly, absorb the visible light in the absorption end of longer wavelength region of the absorption maximum, are remarkably useful as a material for producing a recording layer of high density optical recording media such as DVD-Rs for the coming generation.

Page 23, please amend paragraph 1 as follows:

The styryl dyes of the present invention can be prepared by various procedures. They can be preferably and economically produced by a step of reacting a quaternary ammonium salt of nitrogen atom containing heterocyclic compound having either an active methyl- or active methylene-group with an aldehyde compound. With this method, the styryl dyes of the present invention can be produced in a desired yield by reacting a compound represented by Formula 9 having ϕ_1 and R_1 corresponding to those in Formula 1 with a compound represented by Formula 10 having ϕ_2 corresponding to those in Formula 1.

Page 24, please amend paragraph 1 as follows:

For example, appropriate amounts (usually about equal mols) of compounds represented by Formulae 9 and 10 are placed in a reaction vessel, and the resulting mixture is dissolved in an adequate solvent, if necessary, and then reacted at ambient temperature of over the temperature or under heating and stirring conditions, for example, a reflux in the presence of an appropriate amount of a basic compound such as sodium hydroxide, potassium hydroxide, sodium carbonate, potassium carbonate, sodium hydrogencarbonate, potassium hydrogencarbonate, ammonia, triethylamine, piperidine, pyridine, pyrrolidine, aniline, N,N-

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dimethylaniline, N,N-diethylaniline, or N-methyl-2-pyrrolidone; an acidic compound such as hydrochloric acid, sulfuric acid, nitric acid, methanesulfonic acid, p-toluenesulfonic acid, acetic acid, acetic anhydride, propionic anhydride, trifluoroacetic acid, or trifluorosulfonic acid; or a Lewis acidic compound such as aluminium chloride, zinc chloride, tin tetrachloride, or titanium tetrachloride.

Page 24, please amend paragraph 2 as follows:

As to solvents which can be used in the present invention, for example, hydrocarbons such as pentane, hexane, cyclohexane, octane, benzene, toluene, and xylene; halides such as carbon tetrachloride, chloroform, 1,2-dichloroethane, 1,2-dibromoethane, trichloroethylene, tetrachloroethylene, chlorobenzene, bromobenzene, and 1,4-dichlorobenzene; alcohols and phenols such as methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, isobutyl alcohol, isopentyl alcohol, cyclohexanol, ethylene glycol, propylene glycol, 2-methoxyethanol, 2-ethoxyethanol, phenol, benzyl alcohol, cresol, diethylene glycol, triethylene glycol, and glycerin; ethers such as diethyl ether, diisopropyl ether, tetrahydrofuran, tetrahydropyran, 1,4-dioxane, anisole, 1,2-dimethoxyethane, diethylene glycol dimethyl ether, dicyclohexyl-18-crown-6, methylcarbitol, and ethylcarbitol;

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ketones such as furfural, acetone, ethyl methyl ketone, and cyclohexanone; acids and acidic derivatives such as acetic acid, acetic anhydride, trichloroacetic acid, trifluoroacetic acid, propionic anhydride, ethyl acetate, butyl carbonate, ethylene carbonate, propylene carbonate, formamide, N-methylformamide, N,N-dimethylformamide, N-methylacetoamide, N,N-dimethylacetoamide, hexamethylphosphoric triamide, and trimethyl phosphate; nitriles such as acetonitrile, propionitrile, succinonitrile, and benzonitrile; nitro compounds such as nitromethane and nitrobenzene; sulfur-containing compounds such as dimethylsulfoxide and sulfolane; and water can be used and arbitrarily used in a combination, if necessary.

Page 25, please amend paragraph 1 as follows:

In general, when solvents are used, the greater the volume of solvents the lower the reaction efficiency. On the contrary, the lower the volume of solvents the more difficult homogeneous heating and stirring of the contents becomes and undesirable side reactions may easily occur. Thus, the volume of solvents is desirably set to a level up to 100-folds, usually, in the range of 5-50-folds to the total volume of material compounds by weight. The reaction is complete within 10 hours, usually, 0.5-10 hours, depending on the kinds of reactants and the reaction conditions. The reaction progress can be monitored by

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conventional methods, for example, thin layer chromatography, gas chromatography, and high-performance liquid chromatography. After completion of the reaction, the styryl dyes of the present invention with desired counter ions are obtainable by subjecting the reaction mixture to a conventional counter ion-exchange reaction, if necessary. Thus, all of the styryl dyes represented by Chemical Formulae 1 to 60 are obtainable in a desired yield by the above method. Compounds represented by Formulae 9 and 10 are obtainable in accordance with conventional methods to form a cyclic nucleus in the styryl dyes.

Page 26, please amend paragraph 1 as follows:

The styryl dyes thus obtained can be used in the form of a reaction mixture without any further treatment, but are usually used after being purified by conventional techniques which are generally used for purifying their related compounds, such as dissolution, extraction, separation, decantation, filtration, concentration, thin layer chromatography, column chromatography, gas chromatography, high-performance liquid chromatography, distillation, crystallization, and sublimation. If necessary, these methods can be used in combination. When the styryl dyes of the present invention are used in dye lasers and optical recording media such as DVD-Rs, it is preferable that the styryl dyes of the present invention be distilled, crystallized, and/or sublimated prior to use.

Page 27, please amend paragraph 1 as follows:

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The light absorbents of the present invention comprise one or more of the styryl dyes and include those which utilize the nature of the styryl dyes which substantially absorb visible light with a wavelength around 400 nm. The compositions or the physical forms of the light absorbents are not restricted. Therefore, the light absorbents of the present invention may comprise one or more other compounds in addition to the styryl dyes, depending on use. One of the fields in which the light absorbents of the present invention can be advantageously used is optical recording media. In this field, the light absorbents of the present invention are suitable as a material for producing recording layers of optical recording media using a laser beam with a wavelength of 450 nm or less as a writing light. In using the light absorbents of the present invention in optical recording media, unless the use deviates from the object of the present invention, the styryl dyes can be mixed with other light absorbents comprising other organic dye compounds which are sensitive to visible light and one or more conventional materials used in optical recording media, such as a light-resistant improver, binder, dispersing agent, flame retardant, lubricant, antistatic agent, surfactant, thermal interference inhibitor, plasticizer, color fixing agent, developer, and dissolving assistant, if necessary.

Page 27, please amend paragraph 2 as follows:

Explaining the uses of the light absorbents of the present invention by exemplifying organic optical recording media (organic heat-deformative-type optical recording media), the optical recording media of the present invention can be prepared in accordance with conventional organic recording media because the styryl dyes of the present invention require no special treatment beyond the treatments which are commonly used in this art. For example, one or more of the styryl dyes of the present invention can be mixed with one or more other organic dye compounds which are sensitive to visible light, and one or more conventional light-resistant improvers, binders, dispersing agents, flame retardants, lubricants, antistatic agents, surfactants, thermal interference inhibitors, and plasticizers to control the reflection rate and the light absorption rate in a recording layer, if necessary. The resulting mixture is dissolved in an organic solvent, and the solution is uniformly coated over either surface of the substrates by means of spraying, soaking, roller coating, or rotatory coating methods; and dried to form a recording layer, a thin layer comprising light absorbents, and if necessary, followed by forming a reflection layer to be closely attached onto the recording layer by the method of vacuum deposition, chemical vapor deposition, sputtering, or ion plating using metals such as gold, silver,

In re Appl. KASADA4

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copper, platinum, aluminium, cobalt, tin, nickel, iron, and chromium or using conventional organic materials for reflection layers to impart a reflection efficiency with which the written information can be read, for example, 20% or more, and preferably 30% or more. The ultraviolet-ray-hardening resins or thermosetting resins, which contain flame retardants, stabilizers, and/or antistatic agents are spin coated over the recording layer, and then the coated resins are hardened by either irradiating light or heating to form a protection layer to be closely attached onto the reflection layer to protect the recording layer from scratches, dusts, spills, etc. Thereafter, if necessary, a pair of the above substrates with recording, reflection, and recording layers are faced and attached together using, for example, adhesives or viscous sheets; or protection plates, which are comprising the same materials and shapes as the substrates, are attached to the protection layers of the substrates.

Page 29, please amend paragraph 1 as follows:

As another organic dye compounds usable in combination with the styryl dyes of the present invention, dye compounds are particularly restricted as long as they are sensitive to visible light and can modulate the light reflection- and the light absorption-rates of recording layers of optical recording media. As the organic dye compounds, the following compounds can be

In re Appl. KASADA4

used in an arbitrary combination, if necessary: Acridine dye, azaannulene dye, azo dye, azo metal complex dye, anthraquinone dye, indigo dye, indanthrene dye, oxazine dye, xanthene dye, dioxazine dye, thiazine dye, thioindigo dye, tetrapyrapolphyradine dye, triphenylmethine dye, triphenylthiazine dye, naphthoquinone dye, pyrromethene dye, phthalocyanine dye, benzoquinone dye, benzopyran dye, benzofuranone dye, polyphyrin dye, rhodamine dye, and polymethine dyes such as cyanine dye, merocyanine dye, oxonol dye, azulenium dye, squalilium dye, styryl dye, pyrilium dye, thiopyrilium dye, and phenanthrene dye in which the same or different rings are bound to both ends of polymethine chain such as monomethine, dimethine, trimethine, tetramethine, pentamethine, hexamethine, or heptamethine. The chain and rings can have one or more substituents. Examples of the ring are imidazoline ring, imidazole ring, benzoimidazole ring, α -naphthimidazole ring, β -naphthimidazole ring, indole ring, isoindole ring, indolenine ring, isoindolenine ring, benzoindolenine ring, pyridinoindolenine ring, oxazoline ring, oxazole ring, isooxazole ring, benzooxazole ring, pyridinooxazole ring, α -naphthoxazole ring, β -naphthoxazole ring, selenazoline ring, selenazole ring, benzoselenazole ring, α -naphthselenazole ring, β -naphthselenazole ring, thiazoline ring, thiazole ring, isothiazole ring, benzothiazole ring, α -naphththiazole ring, β -naphththiazole ring, tellurazoline ring, tellurazole ring, benzotellurazole ring, α -

In re Appl. KASADA4

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naphthtellurazole ring, β -naphthtellurazole ring, acridine ring, anthracene ring, isoquinoline ring, isopyrrole ring, imidanoxaline ring, indandione ring, indazole ring, indaline ring, oxadiazole ring, carbazole ring, xanthene ring, quinazoline ring, quinoxaline ring, quinoline ring, chroman ring, cyclohexanedione ring, cyclopentandione ring, cinnoline ring, thiodiazole ring, thiooxazolidone ring, thiophene ring, thionaphthene ring, thiobarbituric acid ring, thiohydantoin ring, tetrazole ring, triazine ring, naphthalene ring, naphthyridine ring, piperazine ring, pyrazine ring, pyrazole ring, pyrazoline ring, pyrazolidine ring, pyrazolone ring, pyran ring, pyridine ring, pyridazine ring, pyrimidine ring, pyrylium ring, pyrrolidine ring, pyrroline ring, pyrrole ring, phenazine ring, phenanthridine ring, phenanthrene ring, phenanthroline ring, phthalazine ring, pteridine ring, furazane ring, furan ring, purine ring, benzene ring, benzoxazine ring, benzopyran ring, morpholine ring, and rhodanine ring. As the organic dye compounds used in combination with the styryl dyes of the present invention, they desirably have an absorption maxima in the visible region, particularly, at wavelengths of 400-850 nm, when formed in a thin layer.

Page 31, please amend paragraph 1 as follows:

The light-resistant improvers are, for example, nitroso compounds such as nitrosodiphenylamine, nitrosoaniline,

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nitrosophenol, and nitrosonaphthol; and metal complexes such as dithiolate metal complexes, for example, "NKX-1199" (bis[2'-chloro-3-methoxy-4-(2-methoxyethoxy)dithiobenzyl]nickel) produced by Hayashibara Biochemical Laboratories, Inc., Okayama, Japan, and formazane metal complexes, which all can be arbitrarily used in combination, if necessary. Preferable light-resistance improvers are those which contain metal complexes, and the most preferable ones are metal complexes, more preferably, formazane compounds which have a pyridine-ring at C-5 and a pyridine or furan ring at C-3 of formazane skeleton as disclosed in Japanese Patent Application No. 163,036/99, titled "Formazane metal complexes", and complexes with metals such as nickel, zinc, cobalt, iron, copper, and palladium, which have the tautomer of the aforesaid compounds as a ligand. In the case of using such a light-resistance improver in combination, the styryl dyes of the present invention can be effectively prevented from undesirable deterioration, fading, color changing, and quality changing, which are inducible by environmental lights such reading- and natural-lights, without lowering the solubility of the light absorbents of the present invention in organic solvents and substantially deteriorating preferable optical features. As to a composition ratio, 0.01-5 moles of a light-resistance improver(s), desirably, 0.1-1 moles, can be incorporated into one mole of the styryl dye(s) while increasing or decreasing the ratio. The light-resistant improver(s) are not necessarily other

In re Appl. KASADA4

compound(s) which exist separately from the styryl dyes of the present invention, and if necessary, they can be formulated into salts, complexes, or compounds by combining the styryl dyes with organic metal complex anions, which are capable of improving the light resistance as disclosed in Japanese Patent Kokai Nos. 19,355/89, 139,043/93, 323,478/97, and 6,651/98. These include conventional compounds such as azo, bisphenyldithiol, phenyldithiol, thiocatecholchelate, thiobisphenolatechelate, or bisdithiol- α -diketone by using appropriate spacers and crosslinking agents such as alkoxides or cyanates of titanium, zirconium, aluminium, etc., or complexes of these metal elements having carbonyl compounds or hydroxy compounds as ligands.

Page 32, please amend paragraph 1 as follows:

The styryl dyes of the present invention have satisfactorily-high solubility in organic solvents without substantially causing undesirable problems in actual use, and do not particularly restrict the types of organic solvents used for coating the light absorbents of the present invention over substrates. Thus, in the preparation of optical recording media according to the present invention, arbitrary organic solvents can be selected from the following conventional ones which are arbitrarily used in combination, if necessary: TFP frequently used to prepare optical recording media and the following organic solvents other than TFP: For example, hydrocarbons such as

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benzene, toluene, and xylene; halides such as carbon tetrachloride, chloroform, 1,2-dichloroethane, 1,2-dibromoethane, trichloroethylene, tetrachloroethylene, chlorobenzene, bromobenzene, and α -dichlorobenzene; alcohols and phenols such as methanol, ethanol, propanol, isopropanol, 2,2,2-trifluoroethanol, butanol, isobutanol, isopentanol, cyclohexanol, ethylene glycol, propylene glycol, 2-methoxyethanol (methyl cellosolve), 2-ethoxyethanol (ethyl cellosolve), phenol, benzyl alcohol, cresol, diethylene glycol, triethylene glycol, glycerine, and diacetone alcohol; ethers such as diethyl ether, diisopropyl ether, tetrahydrofuran, tetrahydropyran, 1,4-dioxane, anisole, 1,2-dimethoxyethane, dicyclohexyl-18-crown-6, methylcarbitol, and ethylcarbitol; ketones such as furfural, acetone, 1,3-diacetylacetone, ethyl methyl ketone, and cyclohexanone; esters such as ethyl acetate, butyl acetate, ethylene carbonate, propylene carbonate, and trimethyl phosphate; amides such as formamide, N-methylformamide, N,N-dimethylformamide, and hexamethylphosphoric triamide; nitriles such as acetonitrile, propionitrile, and succinonitrile; nitro compounds such as nitromethane and nitrobenzene; amines such as ethylene diamine, pyridine, piperidine, morpholine, and N-methylpyrrolidone; and sulfur-containing compounds such as dimethylsulfoxide and sulfolane.

Page 33, please amend paragraph 1 as follows:

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Particularly, since the styryl dyes of the present invention have relatively high solubilities in easily volatile organic solvents such as TFP, methyl cellosolve, ethyl cellosolve, and diacetone alcohol, they are substantially free from dye crystallization when they are sequentially dissolved in the organic solvents, coated on substrates, and dried and do not cause inconsistent thickness and surface of the formed recording layers. Most of the styryl dyes of the present invention exert desirable solubilities in non-halogen solvents, for example, cellosolves such as methyl cellosolve and ethyl cellosolve, alcohols such as diacetone alcohol, and ketones such as ethyl methyl ketone and cyclohexanone. When the styryl dyes of the present invention dissolve in the non-halogen solvents and the obtained solutions are coated over the plastic substrates, the solvents neither damage the substrates nor pollute the environment, which is an advantage.

Page 34, please amend paragraph 1 as follows:

Conventional substrates can be used in the present invention and are usually processed with suitable materials, for example, into discs, 12 cm in diameter and 0.1-1.2 mm in thickness, to be conformed to final use by methods such as compression molding, injection molding, compression-injection molding, photopolymerization method (2P method), thermosetting integral method, and lightsetting integral method. These discs

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can be used singularly or plurally after they have been appropriately attached together with adhesive sheets or adhesive agents, etc. In principal, any materials for substrates can be used in the present invention as long as they are substantially transparent and have light transmissivity of at least 80%, and preferably 90% or more at wavelengths ranging from 350 nm to 800 nm. Examples of such materials are glasses, ceramics, and others such as plastics including polyacrylate, polymethyl methacrylate, polycarbonate, polystyrene (styrene copolymer), polymethylpentene, polyester, polyolefin, polyimide, polyetherimide, polysulfone, polyethersulfone, polyarylate, polycarbonate/polystyrene alloy, polyestercarbonate, polyphthalatecarbonate, polycarbonateacrylate, non-crystalline polyolefin, methacrylate copolymer, diallylcarbonatediethylene-glycol, epoxy resin, and phenol resin, where polycarbonate and acrylate are most frequently used. In plastic substrates, concaves for expression of synchronizing signals and addresses of tracks and sectors are usually transferred to the internal circle of the tracks during their formation. The present invention does not restrict the form of concaves and the concaves are preferably formed to give 0.3-0.8 nm in average width and 50-150 nm in depth.

Page 35, please amend paragraph 1 as follows:

The styryl dyes of the present invention can be

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polyacrylonitrile; polyesters such as poly(ethylene terephthalate); and polyolefins such as polyethylene, chlorinated polyethylene, and polypropylene.

Page 36, please amend paragraph 1 as follows:

Explaining the manner for using the optical recording media according to the present invention, the high-density optical recording media of the present invention such as DVD-Rs can write informations at a relatively high density by using a laser beam with a wavelength of 450 nm or less, more particularly, a laser beam with a wavelength around 350-450 nm irradiated by semiconductor laser elements such as InN, GaN, InGa_N, InAlGa_N, InGa_NAs, BInN, InGaNP, InP, GaP, GaAsP, and SiC, which oscillate in the blue color or violet color region; or a distributed feedback-type laser, which oscillates in the red color region, and to which is set a second harmonics generating mechanism to a semiconductor element of AlGaAs. To read recorded information, laser beams are used with wavelengths identical to or slightly longer or shorter than those used for writing information. As for the laser power for writing and reading information, in the optical recording media of the present invention, it is preferably set to a relatively high level, which exceeds the threshold of the energy required for forming pits in writing information. It is preferably set to a relatively low level, i.e., a level of below the threshold when used in reading

In re Appl. KASADA4

recorded informations, although the power levels can be varied depending on the types and ratios of the light-resistant improvers used in combination with the light absorbents of the present invention and further the writing speed. Generally, the levels can be controlled to outputs of over 5 mW for writing, usually in the range of 10-50 mW, and to outputs of 5 mW or lower for reading, usually in the range of 0.1-5 mW. The recorded information is read by detecting the changes of both the reflection light level and the transmission light level in the pits and the pit-less part on the surface of optical recording media by the light pick-up manner.

Page 37, please amend paragraph 1 as follows:

Accordingly, in the optical recording media of the present invention, remarkably minute pits with a pit width of below 0.4 nm/pit and a track pitch of below 0.74 nm used in the conventional DVD-R, can be formed at a relatively high density by using a laser beam with an oscillating wavelength of 450 nm. For example, in using a substrate, 12 cm in diameter, it can realize an extremely high density optical recording medium with an optical recording capacity far exceeding 4.7 GB (giga bytes) per one side, i.e., that of about two hours of information of sound and images in relatively high image quality of high definition television which could hardly be attained in the commonly used DVD-Rs.

Page 37, please amend paragraph 2 as follows:

Since the optical recording media of the present invention can record information including characters, images, and sound at a relatively high density, they are advantageously useful as recording media for professional and family uses to record and care for documents, data, and computer software. Particular examples of the types of industries and the forms of information to which the optical recording media can be applied are drawings of construction and engineering works, maps, ledgers of roads and rivers, aperture cards, architectural sketches, documents of disaster protection, wiring diagrams, arrangement plans, information from newspapers and magazines, local information, and construction specifications, which all relate to construction and engineering works; blueprints, ingredient tables, prescriptions, product specifications, product price tables, parts lists, information for maintenance, case study files of accidents and problems, manuals for claims, production schemes, technical documents, sketches, details, company's house-made product files, technical reports, and analysis reports, which are all used in production; customers information, information of business connections, information of companies, contracts, information of newspapers and magazines, business reports, company's credit research, and stock lists, which are all used in sales; information of companies, records of stock

In re Appl. KASADA4

prices, statistical documents, information from newspapers and magazines, contracts, customer lists, documents of application/notification/licenses/authorization, and business reports, which are all used in finance; information regarding real property and transportation, sketches of construction, maps, local information, information from newspapers and magazines, contracts for lease, information about companies, stock lists, traffic information, and information regarding business connections, which are all used in real property and transportation; diagrams of writings and pipe arrangements for electric and gas supplies, documents of disaster protection, tables of operation manuals, documents of investigations, and technical reports; medical charts, files of clinical histories and case studies, and diagrams for medical care/institution relationships; texts, collections of questions, educational documents, and statistical information; scientific papers, records in academic societies, monthly reports of research, research data, documentary records and indexes thereof, which are all used in universities, colleges, and research institutes; inspection data, literatures, patent publications, weather maps, analytical records of data, and customer files, which are all used for information; case studies on laws; membership lists, history notes, records of works/products, competition data, and data of meetings/congresses, which are all used in several organizations/associations; sightseeing information, traffic

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information, and local information, which are all used for sightseeing; indexes of homemade publications, information from newspapers and magazines, who's who files, sport records, telop files, and scripts, which are all used in mass communication and publishing; and maps, ledgers of roads and rivers, fingerprint files, resident cards, documents of application/notification/license/authorization, statistical documents, and public documents, which are all used in government offices. Particularly, the write-once type optical recording media of the present invention can be advantageously useful for storing records of charts and official documents which should never be falsified and deleted, and used as electronic libraries for art galleries, libraries, museums, broadcasting stations, etc.

Page 40, please amend paragraph 1 as follows:

As a rather specific use, the optical recording media of the present invention can be used to edit and proofread compact discs, digital video discs, laser discs, MD (a mini disc as an information recording system using photomagnetic disc), CDV (a laser disc using compact disc), DAT (an information recording system using magnetic tape), CD-ROM (a read-only memory using compact disc), DVD-ROM (a read-only memory using digital video disc), DVD-RAM (a writable and readable memory using digital video disc), digital photos , movies, computer graphics,

In re Appl. KASADA4

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compounds via the styryl dyes by using the styryl dyes along with one or more other organic dye compounds which are sensitive to a laser beam with a longer wavelength, e.g., a laser beam with a wavelength around 635-650 nm or 775-795 nm, resulting in a decomposition of the organic dye compounds. The optical recording media as referred to in the present invention mean optical recording media in general which use the characteristics of the specific styryl dyes of the present invention that have an absorption maxima at a wavelength of 400 nm or less and substantially absorb visible light with a wavelength around 400 nm in addition to organic thermal-deformed optical recording media, thermal coloration method using the chemical reaction of coloring agents and developers using the heat generated when organic dye compounds absorb light, and the technique called "moth-eye type technique" which uses the phenomenon that the above heat smooths the pattern of periodical unevenness provided on the surface of the substrates.

Page 41, please amend paragraph 1 as follows:

Since the styryl dyes of the present invention have absorption maxima at a wavelength of 400 nm or less and substantially absorb visible light with a wavelength around 400 nm, the light absorbents of the present invention containing the styryl dyes can be advantageously useful in the aforesaid optical recording media and also used as materials for polymerizing

In re Appl. KASADA4

polymerizable compounds by exposure to visible light, sensitizing solar batteries, light absorptive materials for lithography, materials for laser active substances in dye lasers which oscillate in the blue color or violet region, and for dying clothes in combination with one or more other organic dye compounds which substantially absorb visible light. If necessary, in combination with one or more other light absorbents capable of absorbing light in the ultraviolet, visible and/or infrared regions, the light absorbents can be used in clothes in general and others including building/bedding/decorating products such as drapes, lace, casements, prints, casement cloth, roll screens, shutters, shop curtains, blankets, thick bed quilts including comforters, peripheral materials for the thick bed quilts, cover for the thick bed quilts, cotton for the thick bed quilts, bed sheets, cushions, pillows, pillow covers, cushions, mats, carpets, sleeping bags, tents, interior finish for cars, and window glasses including car window glass; sanitary and health goods such as paper diapers, diaper-covers, eyeglasses, monocles, and lorgnettes; internal base sheets, linings, and materials for shoes; wrappers; materials for umbrellas; parasols; stuffed toys; lighting devices; filters, panels and screens for information displaying devices such as televisions and personal computers which use cathode-ray tubes, liquid crystal displays, electroluminescent displays, and plasma displays; sunglasses; sunroofs; sun visors; pet bottles; refrigerators; vinyl houses;

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lawns; optical fibers; prepaid cards; and windows of ovens including electric ovens. When used as wrapping materials, injection materials, and vessels for the above products, the light absorbents of the present invention prevent living bodies and products from problems and discomforts induced by environmental light such as natural and artificial light or even minimize the above them. Furthermore, they can advantageously regulate the color, tint, and appearance and control the light reflected by or passed through the products to a desirable color balance.

Page 43, please amend paragraph 2 as follows:

Twenty milliliters(ml) of acetic anhydride and 0.6 ml of triethylamine were placed in a reaction vessel, and mixed with 4 g of 2,3,4-trimethylthiazolium=iodide and 2 g of 4-cyanobenzaldehyde. The resulting mixture was reacted at 80°C for one hour under stirring conditions, and then was cooled down.

The crystals formed were collected, washed with ethanol, dissolved in a solution of methanol and chloroform under heating, and then filtered. The obtained filtrate was distilled to remove chloroform and cooled down to obtain 1.1 g of a yellow crystal of the styryl dye represented by Chemical Formula 9. When measured in a conventional manner, the melting point of the crystal was 261-263°C.

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Page 43, please amend paragraph 4 as follows:

Forty milliliters of ethanol and 2 ml of triethylamine were placed in a reaction vessel, and mixed with 6 g of 1,2,3,3-tetramethyl-3H-indolenium tosylate and 3 g of 3-nitrobenzaldehyde. The resulting mixture was reacted at 80°C for one hour under stirring conditions, and then was cooled down. The crystals formed were collected, dissolved in a solution of methanol and chloroform under heating conditions, and then filtered. The obtained filtrate was distilled to remove chloroform and cooled to obtain 3.4 g of an orangish yellow crystal of the styryl dye represented by Chemical Formula 40. When measured in a conventional manner, the melting point of the crystal was 233°C.

Page 45, please amend paragraph 1 as follows:

Ten milliliters of dimethylformamide and 1 ml of iodomethyl were placed in a reaction vessel, and mixed with 1 g of the styryl dye obtained in Example 3. The resulting mixture was reacted at 100°C for one hour under stirring conditions, and then was cooled. The crystals formed were collected and recrystallized in ethanol to obtain 0.8 g of an orangish red crystal of the styryl dye represented by Chemical Formula 36. When measured in a conventional manner, the melting point of the crystal was 222-223°C.

In re Appl. KASADA4

Page 45, please amend paragraph 2 as follows:

Twelve milliliters of ethanol and 0.24 ml of piperidine were placed in a reaction vessel, and mixed with 3 g of 1,3-diethyl-2-methyl-5,6-dichlorobenzimidazolium tosylate and 1.1 g of 4-nitrobenzaldehyde. The resulting mixture was reacted at 80°C for one hour under stirring conditions, and then was cooled. The crystals formed were collected, dissolved in a solution of methanol and water under heating conditions, and then filtered. The obtained filtrate was distilled to concentrate the solution to half volume and cooled to obtain 1.7 g of a yellow crystal of the styryl dye represented by Chemical Formula 23. When measured in a conventional manner, the melting point of the crystal was 287-290°C.

Page 47, please amend paragraph 1 as follows:

As shown in the visible absorption spectra of Fig. 3, the conventional related compound represented by Chemical Formula 61 has absorption maxima at a wavelength longer than 400 nm when formed in a solution and in a thin layer. As shown in the results of Figs. 1 and 2 and Table 1, most of the styryl dyes of the present invention have absorption maxima at a wavelength of 400 nm or less when formed in a solution and in a thin layer, and the absorption ends to the side of longer wavelength area extended to around 450 nm when formed in a thin layer. These

In re Appl. KASADA4

results showed that the styryl dyes of the present invention were different from conventional related compounds and that they were sensitive to a laser beam with a wavelength of 450 nm or less, particularly, a laser beam with a wavelength around 350-450 nm, and substantially absorb such a laser beam.

Page 47, please amend paragraph 2 as follows:

Fifteen milligrams (mg) of either of the styryl dyes of the present invention represented by Chemical Formulae 9, 19, 23, 34, 36, 38, 40, and 44 was added to three milliliters (ml) of TFP. To the mixture was added 2 mg of nitrosodiphenylamine represented by Chemical Formula 62 which is used in conventional optical recording media as a light-resistant improver, followed by 5 min ultrasonic energization at ambient temperature to dissolve the contents in the solvent. Thereafter, in a conventional manner, a prescribed amount of the resulting solution was dropped on either surface of a polished glass substrate, 5 cm × 5 cm, while the glass substrate was rotated at a rotation rate of 1,200 rpm for one minute to uniformly coat the solution thereupon, and sequentially blown with hot air and cold air to form a thin layer on the glass substrate.

Page 48, please amend paragraph 1 as follows:

The resulting glass substrate coated with either of the

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styryl dyes was measured for transmittance (T_0) at a wavelength of 400 nm, and then set to the position seven cm away from a 500 W xenon lamp and exposed to the light of the lamp for 25 min while sending cold air to the substrate. Immediately after that, the resulting substrate was remeasured for transmittance (T) at a wavelength of 400 nm, and the transmittance of T and T_0 for each styryl dye was substituted for the Equation 1 to calculate the residual percentage (%) of the styryl dye. In parallel, control systems with no light-resistant improver for each styryl dye were provided.

Page 51, please amend paragraph 1 as follows:

As shown in the results of Table 2, in the system with no light-resistant improvers, maximum 70% of the styryl dyes had changed with only 25 min exposure of light to be incapable of exerting their inherent light absorbent properties. When used in combination with the nitroso compound represented by Chemical Formula 62, over 76 % of the styryl dyes still remained intact without being changed by the exposure. These results indicate that light-resistant improvers are useful to suppress undesirable changes of the styryl dyes of the present invention by exposure to natural and artificial light.

Page 51, please amend paragraph 2 as follows:

Either of the styryl dyes of the present invention

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represented by Chemical Formulae 8, 9, 19, 23, 34, 36, 38, 40, and 44 as a light absorbent was admixed with TFP to give a concentration of 3.0%(w/w), and the mixture was further added the nitroso compound represented by Chemical Formula 62 to give a concentration of 0.35%(w/w), heated for a while, followed by ultrasonically dissolving the contents. The resulting solution was in a conventional manner filtered through a membrane, coated in a rotatory manner over one side of an acrylic disc substrate, 12 cm in diameter, to which had been transferred concaves for expressing synchronizing signals and addresses of tracks and sectors by an injection molding, to give a thickness of 200 nm. Thereafter, the substrate was spattered with silver to form a reflection layer, 100 nm in thickness, to be closely attached on the surface of the recording layer by vaporization, and the reflection layer was homogeneously coated in a rotatory manner with "DAICURE CLEAR SD1700", a known ultraviolet ray hardening resin commercialized by Dainippon Ink and Chemicals, Inc., Tokyo, Japan, and irradiated to form a protection layer to be closely attached on the surface of the reflection layer, followed by obtaining nine types of optical recording media.

Page 52, please amend paragraph 1 as follows:

All of the optical recording media of this Example can be used for writing large amounts of information in the form of documents, images, and sound at a relatively-high density by

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using a laser element with an oscillation wavelength of 450 nm or less.

Page 52, before paragraph 2 delete Industrial Applicability.

Page 52, please amend paragraph 2 as follows:

As described above, the present invention was made based on the creation of novel styryl dyes and the findings of their industrially usable characteristics. Since the styryl dyes of the present invention have absorption maxima at a wavelength of 400 nm or less and substantially absorb visible light with a wavelength around 400 nm, they have diversified uses in the fields of optical recording media, photochemical polymerization, dye laser, solar batteries, lithography, dyeing, etc., which need organic compounds with such properties. Particularly, among the styryl dyes, they, being sensitive to a laser beam with a wavelength of 450 nm or less, are advantageously useful as a material for a recording layer of high-density optical recording media such as DVD-Rs.

Page 52, please amend paragraph 3 as follows:

As compared with the conventional DVD-Rs which use polymethine dyes as a light absorbent and write information by using a laser beam with a wavelength of 635 nm or 650 nm, the

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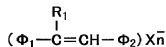
optical recording media of the present invention, which include the styryl dyes of the present invention and use a laser beam with a wavelength of 450 nm or less as a writing light, can form minute pits at a relatively narrower track pitch and a relatively high density. Accordingly, when used the optical recording media of the present invention, they can record a vast amount of information of characters, images, and sound at a relatively-high density, resulting in greatly lowering the cost of recording information per bit and recording animations for a long time, which need much recording volume, of course standing picture as a merit.

IN THE CLAIMS

Please amend claim 1 as follows:

1. (Amended) A styryl dye represented by Formula 1:

Formula 1:



wherein in Formula 1, Φ_1 represents a heterocycle represented by any one of Formulae 2 to 8; Φ_2 represents an optionally substituted aromatic ring or heterocycle; R_1 represents a hydrogen atom, an aliphatic hydrocarbon group, ether group, acyl group, halogen, or cyano group, and the aliphatic hydrocarbon group, ether group, and acyl group may have a substituent; X^- represents a counter ion; and "n" is a number of X^- to balance

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the electric charge in the styryl dye:

Formula 2:



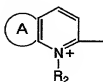
Formula 3:



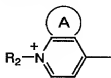
Formula 4:



Formula 5:



Formula 6:



Formula 7:



Formula 8:

In re Appl. KASADA4



throughout Formulae 2 to 7, A represents an optionally substituted monocyclic or polycyclic-aromatic ring or heterocycle; when A is not present in Formulae 2 to 7, one or more substituents similar to those that are bound A may be in the position where A is located; throughout Formulae 2 to 8, R₂ represents an optionally substituted aliphatic hydrocarbon group and R₃ represents a hydrogen or an optionally substituted aliphatic hydrocarbon group which is identical to or different from R₂.

Please amend claim 2 as follows:

2. (Amended) The styryl dye of claim 1, wherein said f_2 is a nitrogen atom containing heterocycle which forms an ammonium salt.

Please amend claim 3 as follows:

3. (Amended) The styryl dye of claim 1, wherein said counter ion is an organic metal complex anion with a light-resistant improving ability.

Please amend claim 4 as follows:

In re Appl. KASADA4

4. (Amended) The styryl dye of claim 1, which has an absorption maximum at a wavelength of 400 nm or less.

Please amend claim 5 as follows:

5. (Amended) The styryl dye of claim 1, which substantially absorbs a visible light with a wavelength of 450 nm or less when formed in a thin layer.

Please amend claim 6 as follows:

6. (Amended) A light absorbent comprising the styryl dye of claim 1.

Please amend claim 7 as follows:

7. (Amended) The light absorbent of claim 6, which further contains one or more other organic dye compounds sensitive to a visible light.

Please amend claim 8 as follows:

8. (Amended) The light absorbent of claim 6, which further contains one or more appropriate light-resistant improvers.

Please amend claim 9 as follows:

9. (Amended) The light absorbent of claim 6, which is sensitive to a visible light with a wavelength of 450 nm

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or less when formed in a thin layer.

Please amend claim 10 as follows:

10. (Amended) An optical recording medium comprising the styryl dye of claim 1.

Please amend claim 11 as follows:

11. (Amended) The optical recording medium of claim 10, which further contains one or more other organic dye compounds sensitive to a visible light.

Please amend claim 12 as follows:

12. (Amended) The optical recording medium of claim 10, which further contains one or more appropriate light-resistant improvers in a recording layer.

Please amend claim 13 as follows:

13. (Amended) The optical recording medium of claim 10, which uses a laser beam with a wavelength of 450 nm or less as a writing light.

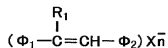
Please amend claim 14 as follows:

14. (Amended) A process for producing the styryl dye

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of

Formula 1:



wherein in Formula 1, Φ_1 represents a heterocycle represented by any one of Formulae 2 to 8; Φ_2 represents an optionally substituted aromatic ring or heterocycle; R_1 represents a hydrogen atom, an aliphatic hydrocarbon group, ether group, acyl group, halogen, or cyano group, and the aliphatic hydrocarbon group, ether group, and acyl group may have a substituent; X^- represents a counter ion; and "n" is a number of X^- to balance the electric charge in the styryl dye:

Formula 2:



Formula 3:

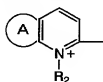


Formula 4:

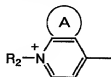


In re Appl. KASADA4

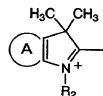
Formula 5:



Formula 6:



Formula 7:



Formula 8:



throughout Formulae 2 to 7, A represents an optionally substituted monocyclic or polycyclic-aromatic ring or heterocycle; when A is not present in Formulae 2 to 7, a substituent similar to A is in the position filled by A; throughout Formulae 2 to 8, R₂ represents an optionally substituted aliphatic hydrocarbon group and R₃ represents a hydrogen or an optionally substituted aliphatic hydrocarbon group which is identical to or different from R₂.

comprising reacting a compound of Formula 9

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Formula 9:



with a compound of Formula 10

Formula 10:



IN THE ABSTRACT

Please enter the new abstract submitted herewith on a separate sheet of paper.

The present invention provides styryl dyes which have absorption maxima at a wavelength of 400 nm or less and are sensitive to a laser beam with a wavelength of 450 nm or less, light absorbents and optical recording media which comprise the styryl dyes, and a process for producing the styryl dyes which comprises reacting with an aldehyde compound a quaternary ammonium salt of nitrogen atom containing heterocyclic compound having an active methyl or active methylene group.

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REMARKS


The present amendment is submitted in order to conform the language of the specification and claims to U.S. practice.

Applicant submits that the invention is new and unobvious and not disclosed by the cited art. Accordingly, Applicant respectfully solicits the Examiner's early review and issuance of this application.

Respectfully submitted,

BROWDY AND NEIMARK, P.L.L.C.
Attorneys for Applicant(s)

By


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AMK:nmp

F:\s\sum\kasada4\pto\prelim amend

ABSTRACT OF THE DISCLOSURE

The present invention provides styryl dyes which have absorption maxima at a wavelength of 400 nm or less and are sensitive to a laser beam with a wavelength of 450 nm or less, light absorbents and optical recording media which comprise the styryl dyes, and a process for producing the styryl dyes which comprises reacting with an aldehyde compound a quaternary ammonium salt of nitrogen atom containing heterocyclic compound having an active methyl or active methylene group.

"Version with markings to show changes"

IN THE SPECIFICATION

Page 1, please amend paragraph 2 as follows:

In a multimedia age, optical recording media such as CD-R (a write-once memory using compact disc) and DVD-R (a write-once memory using digital video disc) are now ~~greatly highlighted~~ of great importance. Most of the conventional optical recording media can be roughly classified into inorganic optical recording media which have recording layers composed of inorganic substances such as tellurium, selenium, rhodium, carbon, or carbon sulfide; and organic optical recording media which have recording layers mainly composed of light absorbents containing organic dye compounds.

Page 1, please amend paragraph 3 as follows:

Among these optical recording media, organic optical recording media can be usually prepared by dissolving a polymethine dye in an organic solvent such as 2,2,3,3-tetrafluoro-1-propanol (abbreviated as "TFP" hereinafter), ~~spin~~ spin coating the solution ~~on~~ onto the surface of a polycarbonate substrate, drying the solution to form a recording layer, and sequentially forming and coating onto the surface of the recording layer a reflection layer comprising a metal such as gold, silver or copper, and a protection layer

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comprising an ultraviolet ray hardening resin. When compared with inorganic optical recording media, organic ones may have the drawback that their recording layers are susceptible to change under environmental lights such as reading- and natural- ~~lights~~ light. Organic optical recording media, however, have the ~~merit~~ advantage that they can be made into optical recording media at a lesser cost because their recording layers can be directly formed by coating light absorbents in solution on the surface of substrates. Further, organic optical recording media composed of organic materials are now mainly used as low-cost optical recording media because they are substantially free of corrosion even when contacted ~~with~~ by moisture or sea water and because information recorded therein in a prescribed format can be read out by using commercially available read-only readers after ~~the establishment of~~ establishing thermal-deformation-type optical recording media, a kind of organic optical recording media.

Page 2, please amend paragraph 1 as follows:

What is urgently required in organic optical recording media is to increase their storage capacity to suit to this multimedia age. The research for such an increment, which is now being eagerly continued in this field, is to

increase ~~a~~ the recording capacity per one side from 4.7 GB (giga bite) to 15 GB or more by shortening the wavelength for writing information to 450 nm or less from the wavelengths of 635-650 nm which are commonly used. The high-density optical recording media have the capacity to record movie and animations for six hours in picture quality of standard television or for two hours even in relatively-high picture quality of high definition television. However, since most ~~of~~ organic dye compounds now used in the conventional optical recording media ~~do not suit to a~~ cannot be used with visible light with a wavelength of 450 nm or less, they could not fulfil the need for high-storage density required in many fields.

Page 3, please amend paragraph 2 as follows:

The present inventors eagerly studied and screened compounds. As a result, they found that specific styryl dyes (may be called "styryl dyes" hereinafter), which are obtainable through a step of reacting a quaternary ammonium salt of nitrogen heterocyclic compound having an active methyl- or active methylene- group with an aldehyde compound, have absorption maxima at a wavelength of 400 nm or less and substantially absorb ~~a~~ visible light with a wavelength around 400 nm. The present inventors confirmed that the styryl dyes, which substantially

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absorb a-visible light with a wavelength of 450 nm or less when formed in a thin layer, form remarkably__-minute pits at a relatively__-high density in optical recording media by irradiating with a laser beam with a wavelength of 450 nm or less. The present invention was made based on the creation of novel styryl dye dyes and the discovery of their industrially useful characteristics.

Page 4, please amend paragraph 1 as follows:

FIG. 1 is the visible absorption spectra of one of the styryl dyes of the present invention when in a solution form and in a thin layer form, respectively.

Page 4, please amend paragraph 2 as follows:

FIG. 2 is the visible absorption spectra of another styryl dye of the present invention when in a solution form and in a thin layer form, respectively.

Page 4, please amend paragraph 3 as follows:

FIG. 3 is the visible absorption spectra of a conventional related compound when in a solution form and in a thin layer form, respectively.

Page 4, please amend paragraph 5 as follows:

In Formula 1, R₁ represents a hydrogen atom, an aliphatic hydrocarbon group, ether group, acyl group, halogen, or cyano group. The aliphatic hydrocarbon group in R₁ represented that having up to six carbon atoms, usually, from one to five carbon atoms such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, *sec*-butyl, *tert*-butyl, pentyl, isopentyl, neopentyl, or *tert*-pentyl group. Examples of the ether groups are methoxy, ethoxy, propoxy, isopropoxy, ~~butoxy~~ butoxy, ~~tert-butoxy~~ butoxy, pentoxy, and phenoxy groups. Examples of the acyl groups are formyl, acetyl, propionyl, butyryl, *tert*-butyryl, benzoyl, *o*-tolyl, *m*-tolyl, and *p*-tolyl groups. Examples of the halogens are fluorine, chlorine, bromine, and iodine. One or more of the hydrogens in the aliphatic hydrocarbon group, ether group, and acyl group may be replaced with a short-chain aliphatic hydrocarbon group such as methyl, ethyl, or propyl group; a short-chain ether group such as methoxy, ethoxy, or propoxy group; or a halogen such as fluorine, chlorine, bromine, or iodine.

Page 6, please amend paragraph 1 as follows:

Throughout Formulae 2 to 7, A represents a monocyclic-or polycyclic-aromatic-ring or heterocycle. Examples of the aromatic ring are benzene- and naphthalene-

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rings. Examples of the heterocycle are pyridine, quinoline, and quinoxaline rings. The aromatic ring and heterocycle may have one or more substituents, for example, a halogen atom such as fluorine, chlorine, bromine, or iodine atom; a short-chain aliphatic hydrocarbon group such as methyl, trifluoromethyl, ethyl, propyl, isopropyl, butyl, isobutyl, *sec*-butyl, or *tert*-butyl group; a short-chain ether group such as methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, butoxy butoxy, or *tert*-~~butoxy~~ butoxy group; an alkylaminosulfonyl group such as methylaminosulfonyl, dimethylaminosulfonyl, ethylaminosulfonyl, diethylaminosulfonyl, propylaminosulfonyl, dipropylaminosulfonyl, butylaminosulfonyl, or dibutylaminosulfonyl group; an aromatic hydrocarbon group such as phenyl, biphenyl, *o*-tolyl, *m*-tolyl, *p*-tolyl, xylyl, mesityl, *o*-cumenyl, *m*-cumenyl, *p*-cumenyl, or naphthyl group; an ester group such as methoxycarbonyl, trifluoromethoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, acetoxyl, trifluoroacetoxyl, or benzoyloxy group; nitro group, cyano group, carboxy group, or sulfo group. When A ~~does not exist~~ is not present in Formulae 2 to 7, one or more substituents similar to ~~that combines for~~ those that are bound to A may be in the position ~~to~~ where A ~~may exist~~ is located.

Page 7, please amend paragraph 1 as follows:

Throughout Formulae 2 to 8, R₂ represents an aliphatic hydrocarbon group selected from those having up to eight carbon atoms, usually, up to six carbon atoms such as methyl, ethyl, propyl, isopropyl, isopropenyl, 1-propenyl, 2-propenyl, 2-propynyl, butyl, isobutyl, *sec*-butyl, *tert*-butyl, 2-butenyl, 2-butylnyl, 1,3-butadienyl, pentyl, isopentyl, neopentyl, 1-methylpentyl, 2-methylpentyl, 2-pentenyl, 2-pentynyl, 2-penten-4-ynyl, hexyl, or isohexyl group. The aliphatic hydrocarbon group may have one or more substituents, for example, a halogen such as fluorine, chlorine, bromine, or iodine; an ether group such as methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, ~~buthoxy~~ butoxy, ~~tert-buthoxy~~ butoxy, benzyloxy, or phenoxy group; an ester group such as methoxycarbonyl, trifluoromethoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, acetoxyl, trifluoroacetoxyl, or benzoyloxy group; a carboxy group; a sulfo group; a hydroxy group; an amido group such as acetyl amino or benzoyl amino group; an aromatic hydrocarbon group such as phenyl, biphenyl, o-tolyl, m-tolyl, p-tolyl, xylyl, mesityl, o-cumenyl, m-cumenyl, p-cumenyl, or naphthyl group; or heterocyclic group such as 2-pyridyl, 2-quinolyl, 2-tetrahydropyranyl, 2,2-dimethyl-1,3-dioxolane-4-yl, 1,3-dioxolane-2-yl, 3,5-dimethylisooxazole-4-yl, 3-piperidinyl, piperidino, morpholino, 1-piperazinyl, pyrrolidine-1-yl, 1-methyl-2-pyrrolidinyl, 2-benzimidazolyl, 5-

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Page 8, please amend paragraph 1 as follows:

Page 8, please amend paragraph 2 as follows:

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ring and heterocycle may have one or more substituents. The substituents are selected from those having up to eight carbon atoms, usually, one to six carbon atoms, for example, an aliphatic hydrocarbon group such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, *sec*-butyl, *tert*-butyl, pentyl, isopentyl, neopentyl, *tert*-pentyl, 1-methylpentyl, 2-methylpentyl, hexyl, or isohexyl group; a short-chain ether group such as methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, butoxy butoxy, or *tert*-butoxy butoxy group; an amido group such as acetylamino or benzoylamino group; a halogen such as fluorine, chlorine, bromine, or iodine; and cyano group, nitro group, hydroxy group, sulfo group, or carboxy group. When ϕ_2 is a nitrogen-atom-containing heterocycle, the nitrogen atom may combine with ~~the a~~ similar aliphatic hydrocarbon group as in R_2 in ϕ_1 to form an ammonium salt. When ϕ_2 has two hydroxy groups as a substituent and the hydroxy groups combine with adjacent carbon atoms in ϕ_2 , the hydroxy groups may react with an oxo compound such as formalin to form a cyclic structure such as a dioxole ring.

Page 9, please amend paragraph 1 as follows:

X⁻ in Formula 1 represents an arbitrary counter ion and is usually selected from inorganic acid ions such as fluoric fluoride, chloric chloride, bromic bromide, iodic

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maxima at a wavelength of 400 nm or less and substantially absorb a visible light with a wavelength around 400 nm, they have diversified uses in the fields, which need organic compounds that absorb the aforesaid visible light. Among the styryl dyes, those, which substantially absorb a visible light with a wavelength of 450 nm or less when formed in a thin layer, particularly, absorb the visible light in the absorption end of longer wavelength region of the absorption maximum, are remarkably useful as a material for producing a recording layer of high--density--optical recording media such as DVD-Rs for the coming generation.

Page 23, please amend paragraph 1 as follows:

The styryl dyes of the present invention can be prepared by various ~~manner~~procedures. They can be preferably and economically produced ~~through-by~~ a step of reacting a quaternary ammonium salt of nitrogen--atom--containing heterocyclic compound having either an active methyl- or active methylene-group with an aldehyde compound ~~from an economical view point~~. With ~~the manner~~this method, the styryl dyes of the present invention can be produced in a desired yield by reacting a compound represented by Formula 9 having ϕ_1 and R_1 corresponding to those in Formula 1 with a compound represented by Formula 10 having ϕ_2 corresponding to those in Formula 1.

Page 24, please amend paragraph 1 as follows:

For example, appropriate amounts (usually about equal mols) of compounds represented by Formulae 9 and 10 are placed in a reaction vessel, and the resulting mixture is dissolved in an adequate solvent, if necessary, and then reacted at ambient temperature of over the temperature or ~~over~~ under heating and stirring conditions, for example, a reflux in the presence of an appropriate amount of a basic compound such as sodium hydroxide, potassium hydroxide, sodium carbonate, potassium carbonate, sodium hydrogencarbonate, potassium hydrogencarbonate, ammonia, triethylamine, piperidine, pyridine, pyrrolidine, aniline, N,N-dimethylaniline, N,N-diethylaniline, or N-methyl-2-pyrrolidone; an acidic compound such as hydrochloric acid, sulfuric acid, nitric acid, methanesulfonic acid, p-toluenesulfonic acid, acetic acid, acetic anhydride, propionic anhydride, trifluoroacetic acid, or trifluorosulfonic acid; or a Lewis acidic compound such as aluminium chloride, zinc chloride, tin tetrachloride, or titanium tetrachloride.

Page 24, please amend paragraph 2 as follows:

As to solvents which can be used in the present invention, for example, hydrocarbons such as pentane, hexane, cyclohexane, octane, benzene, toluene, and xylene; halides such

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Page 25, please amend paragraph 1 as follows:

In general, when solvents are used, the greater the volume of solvents the lower the reaction efficiency₇. ~~On~~ On the contrary, the lower the volume of solvents the more difficult₇ homogeneous heating and stirring of the contents becomes and undesirable side ~~reaction-reactions~~ may easily occur. Thus, the volume of solvents is desirably set to a level up to 100-folds, usually, in the range of 5-50-folds to the total volume of material compounds by weight. The reaction ~~is completes-complete~~ within 10 hours, usually, 0.5-10 hours, depending on the kinds of ~~material-compounds~~ reactants and the reaction conditions. The reaction progress can be monitored by conventional methods, for example, thin layer chromatography, gas chromatography, and high-performance liquid chromatography. After completion of the reaction, the styryl dyes of the present invention with desired counter ions are obtainable by ~~providing-subjecting~~ the reaction mixture to a conventional counter ion-exchange reaction, if necessary. Thus, all of the styryl dyes represented by Chemical Formulae 1 to 60 are obtainable in a desired yield by the above method. Compounds represented by Formulae 9 and 10 are obtainable in accordance with conventional methods to form a cyclic ~~nuclear-nucleus~~ in the styryl dyes.

Page 26, please amend paragraph 1 as follows:

The styryl dyes thus obtained can be used in the form of a reaction mixture without any further treatment, but are usually used after being purified by conventional manners, techniques which are generally used for purifying their related compounds, such as dissolution, extraction, separation, decantation, filtration, concentration, thin layer chromatography, column chromatography, gas chromatography, high-performance liquid chromatography, distillation, crystallization, and sublimation. If necessary, these methods can be used in combination. ~~In using~~When the styryl dyes of the present invention are used in dye lasers and optical recording media such as DVD-Rs, it is preferable that the styryl dyes of the present invention ~~are be~~ distilled, crystallized, and/or sublimated prior to use.

Page 27, please amend paragraph 1 as follows:

The light absorbents of the present invention comprise one or more of the styryl dyes and include those which utilize the nature of the styryl dyes which substantially absorb visible light with a wavelength around 400 nm. The compositions or the physical forms of the light absorbents are not restricted. Therefore, the light absorbents of the present invention may comprise one or more other ~~elements compounds~~ in addition to the styryl dyes, depending on use. One of the fields, in which the

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light absorbents of the present invention can be advantageously used, is ~~of~~-optical recording media. In this field, the light absorbents of the present invention are suitable as a material producing for recording layers of optical recording media using a laser beam with a wavelength of 450 nm or less as a writing light. In using the light absorbents of the present invention in optical recording media, unless the use deviates from the object of the present invention, the styryl dyes can be mixed with other light absorbents comprising other organic dye compounds which are sensitive to visible light and one or more conventional materials used in optical recording media, such as a light-resistant improver, binder, dispersing agent, flame retardant, lubricant, antistatic agent, surfactant, thermal interference inhibitor, plasticizer, color fixing agent, developer, and dissolving assistant, if necessary.

Page 27, please amend paragraph 2 as follows:

Explaining the uses of the light absorbents of the present invention by exemplifying organic optical recording media (organic heat-deformative-type optical recording media), the optical recording media of the present invention can be prepared in accordance with conventional organic recording media because the styryl dyes of the present invention require no special treatment ~~and step beyond the treatments~~ which are commonly used

in this art. For example, one or more of the styryl dyes of the present invention can be mixed with one or more other organic dye compounds which are sensitive to visible light, and one or more conventional light-resistant improvers, binders, dispersing agents, flame retardants, lubricants, antistatic agents, surfactants, thermal interference ~~inhibitor~~inhibitors, and plasticizers to control the reflection rate and the light absorption rate in a recording layer, if necessary. The resulting mixture is dissolved in an organic solvent, and the solution is uniformly coated over either surface of the substrates by means of spraying, soaking, roller coating, or rotatory coating ~~method~~methods; and dried to form a recording layer, a thin layer comprising light absorbents, and if necessary, followed by forming a reflection layer to be closely attached onto the recording layer by the method of vacuum deposition, chemical vapor deposition, sputtering, or ion plating using metals such as gold, silver, copper, platinum, aluminium, cobalt, tin, nickel, iron, and chromium or using conventional organic materials for reflection layers to impart a reflection efficiency with which the written information can be read, for example, 20% or more, and preferably 30% or more, ~~and spun coating over the recording layer.~~ The ultraviolet-ray-hardening resins or thermosetting resins, which contain flame retardants, stabilizers, and/or antistatic agents are spin coated over the

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recording layer, and then ~~hardening~~—the coated resins are
hardened resins—by either irradiating light or heating to form a
protection layer to be closely attached onto the reflection layer
to protect the recording layer from scratches, dusts, ~~spoils~~
spills, etc. Thereafter, if necessary, a pair of the above
substrates with recording-, reflection-, and recording-_layers
are faced and attached together using, for example, adhesives or
viscous sheets; or protection plates, which are comprising the
same materials and shapes as the substrates, are attached to the
protection layers of the substrates.

Page 29, please amend paragraph 1 as follows:

As another organic dye compounds usable in combination
with the styryl dyes of the present invention, dye compounds are
particularly restricted as long as they are sensitive to visible
light and can modulate the light reflection- and the light
absorption-rates of recording layers of optical recording media.

As the organic dye compounds, the following compounds can be
used in an arbitrary combination, if necessary: Acridine dye,
azaannulene dye, azo dye, azo metal complex dye, anthraquinone
dye, indigo dye, indanthrene dye, oxazine dye, xanthene dye,
dioxazine dye, thiazine dye, thioindigo dye,
tetrapyrropolphyradine dye, triphenylmethine dye,
triphenylthiazine dye, naphthoquinone dye, pyrromethene dye,

phthalocyanine dye, benzoquinone dye, benzopyran dye, benzofuranone dye, polyphyrin dye, rhodamine dye, and polymethine dye dyes such as cyanine dye, merocyanine dye, oxonol dye, azulenium dye, squalilium dye, styryl dye, pyririum dye, thiopyririum dye, and phenanthrene dye in which the same or different rings are bound to the both ends of polymethine chain such as monomethine, dimethine, trimethine, tetramethine, pentamethine, hexamethine, or heptamethine. The chain and rings can have one or more substituents. Examples of the ring are imidazolin-imidazoline ring, imidazole ring, banzoimidazole ring, α -naphthimidazole ring, β -naphthimidazole ring, indole ring, isoindole ring, indolenine ring, isoindolenine ring, benzoindolenine ring, pyridinoindolenine ring, oxazoline ring, oxazole ring, isooxazole ring, benzooxazole ring, pyridinooxazole ring, α -naphthoxazole ring, β -naphthoxazole ring, selenazoline ring, selenazole ring, benzoselenazole ring, α -naphthselenazole ring, β -naphthselenazole ring, thiazoline ring, thiazole ring, isothiazole ring, benzothiazole ring, α -naphththiazole ring, β -naphththiazole ring, tellurazoline ring, tellurazole ring, benzotellurazole ring, α -naphthhtellurazole ring, β -naphthhtellurazole ring, acridine ring, anthracene ring, isoquinoline ring, isopyrrole ring, imidanoxaline ring, indandione ring, indazole ring, indaline ring, oxadiazole ring, carbazole ring, xanthene ring, quinazoline ring, quinoxaline

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ring, quinoline ring, chroman ring, ~~cyclohexanedien~~
cyclohexanedione ring, ~~cyclopentandien~~ cyclopentandion ring,
cinnoline ring, thiodiazole ring, thiooxazolidone ring, thiophene
ring, thionaphthene ring, thiobarbituric acid ring, thiohydantoin
ring, tetrazole ring, triazine ring, naphthalene ring,
naphthyridine ring, piperazine ring, pyrazine ring, pyrazole
ring, pyrazoline ring, pyrazolidine ring, pyrazolone ring, pyran
ring, pyridine ring, pyridazine ring, pyrimidine ring, pyrylium
ring, pyrrolidine ring, pyrroline ring, pyrrole ring, phenazine
ring, phenanthridine ring, phenanthrene ring, phenanthroline
ring, phthalazine ring, pteridine ring, furazane ring, furan
ring, purine ring, benzene ring, benzoxazine ring, benzopyran
ring, morpholine ring, and rhodanine ring. As the organic dye
compounds used in combination with the styryl dyes of the present
invention, they ~~are-desirable-todesirably~~ have ~~the-an~~ absorption
maxima in the visible region, particularly, at wavelengths of
400-850 nm, when formed in a thin layer.

Page 31, please amend paragraph 1 as follows:

The light-resistant improvers are, for example,
nitroso compounds such as nitrosodiphenylamine, nitrosoaniline,
nitrosophenol, and nitrosonaphthol; and metal complexes such as
dithiolate metal complexes, for example, "NKX-1199" (bis[2'-

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compound(s) which ~~are-exist~~ separately ~~exist~~ from the styryl dyes of the present invention, and if necessary, they can be formulated into salts, complexes, or compounds by combining the styryl dyes with organic metal complex anions, which are capable of improving the light resistance as disclosed in Japanese Patent Kokai Nos. 19,355/89, 139,043/93, 323,478/97, and 6,651/98, ~~such as. These include~~ conventional ~~these-of~~ compounds such as azo, bisphenyldithiol, phenyldithiol, thiocatecholchelate, thiobisphenolatechelate, or bisdithiol- α -diketone by using appropriate spacers and crosslinking agents such as alkoxides or ~~eyarates-cyanates~~ of titanium, zirconium, aluminium, etc., or complexes of these metal elements having carbonyl compounds or hydroxy compounds as ligands.

Page 32, please amend paragraph 3 as follows:

The styryl dyes of the present invention have satisfactorily-high solubility in organic solvents without substantially causing undesirable problems in actual use, and do not particularly restrict the types of organic solvents used for coating the light absorbents of the present invention over substrates. Thus, in the preparation of optical recording media according to the present invention, arbitrary organic solvents can be selected from the following conventional ones which are arbitrarily used in combination,

if necessary: TFP frequently used to prepare optical recording media and the following organic solvents other than TFP: For example, hydrocarbons such as benzene, toluene, and xylene; halides such as carbon tetrachloride, chloroform, 1,2-dichloroethane, 1,2-dibromoethane, trichloroethylene, tetrachloroethylene, ~~chloroene~~chlorobenzene, bromobenzene, and α -dichlorobenzene; alcohols and phenols such as methanol, ethanol, propanol, isopropanol, 2,2,2-trifluoroethanol, butanol, isobutanol, isopentanol, cyclohexanol, ethylene glycol, propylene glycol, 2-methoxyethanol (methyl cellosolve), 2-ethoxyethanol (ethyl cellosolve), phenol, benzyl alcohol, cresol, diethylene glycol, triethylene glycol, glycerine, and diacetone alcohol; ethers such as diethyl ether, diisopropyl ether, tetrahydrofuran, tetrahydropyran, 1,4-dioxane, anisole, 1,2-dimethoxyethane, dicyclohexyl-18-crown-6, methylcarbitol, and ethylcarbitol; ketones such as furfural, acetone, 1,3-diacetylacetone, ethyl methyl ketone, and cyclohexanone; esters such as ethyl acetate, butyl acetate, ethylene carbonate, propylene carbonate, and trimethyl phosphate; amides such as formamide, N-methylformamide, N,N-dimethylformamide, and hexamethylphosphoric triamide; nitriles such as acetonitrile, propionitrile, and succinonitrile; nitro compounds such as nitromethane and nitrobenzene; amines such as ethylene

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diamine, pyridine, piperidine, morpholine, and N-methylpyrrolidone; and sulfur-containing compounds such as dimethylsulfoxide and sulfolane.

Page 33, please amend paragraph 1 as follows:

Particularly, since the styryl dyes of the present invention have relatively high solubilities in easily volatile organic solvents such as TFP, methyl cellosolve, ethyl cellosolve, and diacetone alcohol, they are substantially free from dye crystallization when they are sequentially dissolved in the organic solvents, coated on substrates, and dried, and do not cause inconsistent thickness and surface of the formed recording layers. Most of the styryl dyes of the present invention exert desirable solubilities in non-halogen solvents, for example, cellosolves such as methyl cellosolve and ethyl cellosolve, alcohols such as diacetone alcohol, and ketones such as ethyl methyl ~~keton~~ ketone and cyclohexanone. When the styryl dyes of the present invention dissolve in the non-halogen solvents and the obtained solutions are coated over the plastic substrates, the solvents neither damage the substrates nor pollute the environment ~~as a merit~~, which is an advantage.

Page 34, please amend paragraph 1 as follows:

Conventional substrates can be used in the present

invention and are usually processed with suitable materials, for example, into discs, 12 cm in diameter and 0.1-1.2 mm in thickness, to suit-be conformed to final use by ~~the~~ methods such as compression molding, injection molding, compression-injection molding, photopolymerization method (2P method), thermosetting integral method, and lightsetting integral method. These discs can be used singularly or plurally after they have been appropriately attached ~~them~~-together with adhesive sheets or adhesive agents, etc. In principal, any materials for substrates can be used in the present invention as long as they are substantially transparent and have light transmissivity of at least 80%, and preferably 90% or more at ~~the~~-wavelengths ranging from 350 nm to 800 nm. Examples of such materials are glasses, ceramics, and others such as plastics including polyacrylate, polymethyl methacrylate, polycarbonate, polystyrene (styrene copolymer), polymethylpentenepolymethylpentene, polyester, polyolefin, polyimide, polyetherimide, polysulfone, polyethersulfone, polyarylate, polycarbonate/polystyrene alloy, polyestercarbonate, polyphthalatecarbonate, polycarbonateacrylate, non-crystalline polyolefin, methacrylate copolymer, diallylcarbonatediethylene-glycol, epoxy resin, and phenol resin, where polycarbonate and acrylate are most frequently used. In plastic substrates, concaves for expression of synchronizing signals and addresses of tracks and sectors are

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usually transferred to the internal circle of the tracks during their formation. The present invention does not restrict the form of concaves and the concaves are preferably formed to give 0.3-0.8 nm in average ~~wide-width~~ and 50-150 nm in depth.

Page 35, please amend paragraph 1 as follows:

The styryl dyes of the present invention can be prepared into 0.5-5% (w/w) solutions of the organic solvents as mentioned above while considering the viscosity of the solutions, and then uniformly coated over a substrate to form a recording layer ~~with-of~~ 10-1,000 nm, preferably, 50-300 nm in thickness. Prior to the coating, a preliminary layer can be formed over the substrate to improve the protection and the adhesion ability of the substrate, if necessary. Materials of the preliminary layer are, for example, high-molecular substances such as ionomer resins, polyamide resins, vinyl resins, natural resins, silicon, and liquid rubbers. In the case of using binders, the following polymers can be used alone or in combination in a weight ratio of 0.01-10 times of the light absorbent(s): Cellulose esters such as nitrocellulose, cellulose phosphate, cellulose sulfate, cellulose acetate, cellulose propionate, cellulose lactate, cellulose palmitate, and cellulose acetate/propionate; cellulose ethers such as methyl cellulose, ethyl cellulose,

propyl cellulose, and butyl cellulose; vinyl resins such as polystyrene, poly(vinyl chloride), poly(vinyl acetate), poly(vinyl acetal), poly(vinyl butyral), poly(vinyl formal), poly(vinyl alcohol), and poly(vinyl pyrrolidone); copolymer resins such as styrene-butadiene copolymers, styrene-acrylonitrile copolymers, styrene-butadiene-acrylonitrile copolymers, vinyl chloride-vinyl acetate copolymers, and maleic anhydride copolymers; acrylic resins such as poly(methyl methacrylate), poly(methyl acrylate), polyacrylate, polymethacrylate, polyacrylamide, and polyacrylonitrile; polyesters such as poly(ethylene terephthalate); and polyolefins such as polyethylene, chlorinated polyethylene, and polypropylene.

Page 36, please amend paragraph 1 as follows:

Explaining the manner for using the optical recording media according to the present invention, the high-density optical recording media of the present invention such as DVD-Rs can write informations at a relatively-high density by using a laser beam with a wavelength of 450 nm or less, more particularly, a laser beam with a wavelength around 350-450 nm irradiated by semiconductor laser elements such as ~~these of~~-InN, GaN, InGaN, InAlGaN, InGaNAs, BInN, InGaN_P, InP, GaP, GaAsP, and SiC₂ which oscillate in the blue--color_ or violaceous-violet

color region; or a distributed feedback-type__laser, which oscillates in the red-_color region, and to which is set a second harmonics generating mechanism to a semiconductor element of AlGaAs. To read recorded informations, laser beams are used with wavelengths identical to or slightly longer or shorter than those used for writing informationsinformation. As for the laser power for writing and reading informationsinformation, in the optical recording media of the present invention, it is preferably be-set to a relatively-high level, which exceeds the threshold of the energy required for forming pits, in writing information, ~~while it.~~ It is preferably be-set to a relatively-low level, i.e., a level of below the threshold when used in reading recorded informations, although the power levels can be varied depending on the types and ratios of the light-resistant improvers used in combination with the light absorbents of the present invention and further the writing speed. Generally, the levels can be controlled to outputs of over 5 mW for writing, usually in the range of 10-50 mW, and to outputs of 5 mW or lower for reading, usually in the range of 0.1-5 mW. The recorded informations areinformation is read by detecting the changes of both the reflection light level and the transmission light level in the pits and the pit-less part on the surface of optical recording media by the light pick-up manner.

Page 37, please amend paragraph 1 as follows:

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~~Accordingly, in the optical recording media of the~~
present invention, remarkably-minute pits with a pit width of below 0.4 nm/pit and a track pitch of below 0.74 nm used in the conventional DVD-R, can be formed at a relatively-high density by using a laser beam with an oscillating wavelength of 450 nm. For example, in using a substrate, 12 cm in diameter, it can realize an extremely-high density optical recording medium with an optical recording capacity far exceeding 4.7 GB (giga bytes) per one side, i.e., that of about two hours of information of sound and images in relatively-high image quality of high definition television which could hardly be attained in the commonly used DVD-Rs.

Page 37, please amend paragraph 2 as follows:

Since the optical recording media of the present invention can record information ~~of~~-including characters, images, and sound at a relatively-high density, they are advantageously useful as recording media for professional and family uses to record and care for documents, data, and computer software. Particular examples of the types of industries and the forms of information to which the optical recording media can be applied are drawings of construction and engineering works, maps, ledgers

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of ~~leads~~roads and rivers, aperture cards, architectural sketches, documents of disaster protection, wiring diagrams, arrangement plans, ~~informations-of~~information from newspapers and magazines, local information, and construction specifications, which all relate to construction and engineering works; blueprints, ingredient tables, prescriptions, product specifications, product price tables, parts lists, information for maintenance, case study files of accidents and problems, manuals for claims, production schemes, technical documents, sketches, details, company's house-made product files, technical reports, and analysis reports, which are all used in ~~productions~~production; customers information, ~~informations~~information of business connections, ~~informations-information~~information of companies, contracts, ~~informations-of~~information from newspapers and magazines, business reports, company's credit research, and stock lists, which are all used in sales; information of companies, records of stock prices, statistical documents, informations of newspapers and magazines, contracts, ~~customers~~customer lists, documents of application/notification/licenses/authorization, and business reports, which are all used in ~~money~~finance; information ~~of~~regarding real property and ~~transportation~~transportation, sketches of construction, maps, local information, ~~informations~~of information from newspapers and magazines, contracts for lease,

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~~informations-of~~information about companies, stock lists, traffic
~~informations~~information, and ~~informations-of~~information regarding
business connections, which are all used in real property and
~~transportation~~transportation; diagrams of writings and pipe
arrangements for electric and gas supplies, documents of disaster
protection, tables of operation manuals, documents of
investigations, and technical reports; medical ~~cartes~~charts,
files of clinical histories and case studies, and diagrams for
medical care/institution relationships; texts, collections of
questions, educational documents, and statistical information;
scientific papers, records in academic societies, monthly reports
of research, research data, documentary records and indexes
thereof, which are all used in universities, colleges, and
research institutes; inspection data, literatures, patent
publications, weather maps, analytical records of data, and
~~customers~~customer files, which are all used for information;
case studies on laws; membership lists, history notes, records of
works/products, competition data, and data of
meetings/congresses, which are all used in several
organizations/associations; sightseeing information, traffic
information, and local information, which are all used for
sightseeing; indexes of homemade publications, ~~information of~~
from newspapers and magazines, who's who files, sport records,
telop files, and scripts, which are all used in mass

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~~communications-communication~~ and publishing; and maps, ledgers of roads and rivers, fingerprint files, resident cards, documents of application/notification/license/authorization, statistical documents, and public documents, which are all used in government offices. Particularly, the write-once type optical recording media of the present invention can be advantageously useful for storing records of charts and official documents which should never be falsified and deleted, and used as electronic libraries for art galleries, libraries, museums, broadcasting stations, etc.

Page 40, please amend paragraph 1 as follows:

As a rather specific use, the optical recording media of the present invention can be used to edit and proofread compact discs, digital video discs, laser discs, MD (a mini disc as an information recording system using photomagnetic disc), CDV (a laser disc using compact disc), DAT (an information recording system using magnetic tape), CD-ROM (a read-only memory using compact disc), DVD-ROM (a read-only memory using digital video disc), DVD-RAM (a writable and readable memory using digital video disc), digital photos , movies, computer graphics, publishing products, broadcasting programs, commercial messages, computer programs, video software, audio software, game software, etc.; and used as external program recording means for large size

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~~of~~-computers and car navigation systems.

Page 40, please amend paragraph 1 as follows:

Hereinbefore, the use of the light absorbents of the present invention in the field of optical recording media has been mainly ~~explained with reference to their application~~ examples limited to high-density optical recording media which use a laser beam with a wavelength of 450 nm or less as a writing light. However, in the field of optical recording media, the light absorbents of the present invention can be also advantageously used as materials for changing or regulating the optical absorption rate or the optical reflection rate in the exisiting optical recording media such as CD-Rs, DVD-Rs, and other high-density optical recording media by using in combination, for example, with one or more other organic dye compounds which are sensitive to a laser beam with a wavelength around 635-650 nm or 775-795 nm. When applied to organic-ablation type optical recording media using a laser beam with a wavelength of 450 nm or less as a writing light, the styryl dyes of the present invention can be used not to directly form pits on substrates but to indirectly form pits in such a manner that the excitation energy of a laser beam with a wavelength of 450 nm or less is allowed to transfer to the aforesaid organic dye

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compounds via the styryl dyes by using the styryl dyes along with one or more other organic dye compounds which are sensitive to a laser beam with a longer wavelength, e.g., a laser beam with a wavelength around 635-650 nm or 775-795 nm, resulting in a decomposition of the organic dye compounds. The optical recording media as referred to in the present invention mean optical recording media in general which use the characteristics of the specific styryl dyes of the present invention that have ~~the-an~~ absorption maxima at a wavelength of 400 nm or less and substantially absorb a-visible light with a wavelength around 400 nm in addition to organic thermal-deformed optical recording media, thermal coloration method using the chemical reaction of coloring agents and developers using the heat generated when organic dye compounds absorb light, and the technique called "moth-eye type technique" which uses the phenomenon ~~of~~-that the above heat ~~smoothes-smooths~~ the pattern of periodical unevenness provided on the surface of the substrates.

Page 41, please amend paragraph 1 as follows:

Since the styryl dyes of the present invention have absorption maxima at a wavelength of 400 nm or less and substantially absorb a-visible light with a wavelength around 400 nm, the light absorbents of the present invention containing the styryl dyes can be advantageously useful ~~to-in~~ the aforesaid

optical recording media and also used as materials for polymerizing polymerizable compounds by exposure to visible light, sensitizing solar batteries, light absorptive materials for lithography, materials for laser active substances in dye lasers which oscillate in the blue-color- or ~~violaceous-color~~ violet region, and for dyeing clothes in combination with one or more other organic dye compounds which ~~suit to uses to~~ substantially absorb ~~a~~-visible light. If necessary, in combination with one or more other light absorbents capable of absorbing light in the ultraviolet, visible and/or infrared regions, the light absorbents can be used in clothes in general and others including building/bedding/decorating products such as ~~a drapedrapes~~, lace, ~~casementcasements~~, ~~printprints~~, casement cloth, roll ~~screenscreens~~, ~~shuttersshutters~~, shop ~~curtaincurtains~~, ~~blanketblankets~~, thick ~~bedquilt~~ bed quilts including ~~comfortercomforters~~, peripheral ~~material-materials~~ for the thick ~~bedquiltbed quilts~~, ~~cover-covers~~ for the thick ~~bedquiltbed quilts~~, cotton for the thick ~~bedquiltbed quilts~~, bed ~~sheetssheets~~, ~~cushioncushions~~, ~~pillowpillows~~, pillow ~~covercovers~~, ~~cushioncushions~~, ~~matmats~~, ~~carpetcarpets~~, sleeping ~~bagbags~~, ~~tenttents~~, interior finish for ~~earcars~~, and window glasses including car window glass; sanitary and health goods such as a paper ~~diaperdiapers~~, diaper-covers, eyeglasses, ~~monoclemonocles~~,

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and ~~lorgnettelorgnettes~~; internal base sheets, linings, and materials for shoes; wrappers; materials for umbrellas; parasols; stuffed toys; lighting devices; filters, panels and screens for information displaying devices such as televisions and personal computers which use cathode-ray tubes, liquid crystal displays, electroluminescent displays, and plasma displays; sunglasses; sunroofs; sun visors; pet bottles; refrigerators; vinyl houses; lawns; optical fibers; prepaid cards; and windows of ovens including electric ovens. When used as wrapping materials, injection materials, and vessels for the above products, the light absorbents of the present invention prevent living bodies and products from problems and discomforts induced by environmental ~~lights-light~~ such as natural- and artificial-~~lights~~ light or even minimize the above them. Furthermore, they can advantageously regulate the color, tint, and appearance and control the light reflected by or passed through the products to a desirable color balance.

Page 43, please amend paragraph 2 as follows:

Twenty milliliters(ml) of acetic anhydride and 0.6 ml of triethylamine were placed in a reaction vessel, and mixed with 4 g of 2,3,4-trimethylthiazolium=iodide and 2 g of 4-cyanobenzaldehyde. The resulting mixture was reacted at 80°C for one hour under stirring conditions, and then was

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cooled down. The crystals formed were collected, washed ~~by~~
with ethanol, dissolved in a solution of methanol and
chloroform under heating-~~conditions~~, and then
~~filtrated~~filtered. The obtained filtrate was distilled to
remove chloroform and cooled down to obtain 1.1 g of a yellow
crystal of the styryl dye represented by Chemical Formula 9.
When measured in a conventional manner, the melting point of
the crystal was 261-263°C.

Page 43, please amend paragraph 4 as follows:

Forty milliliters of ethanol and 2 ml of triethylamine
were placed in a reaction vessel, and mixed with 6 g of 1,2,3,3-
tetramethyl-3H-indolenium tosylate and 3 g of 3-
nitrobenzaldehyde. The resulting mixture was reacted at 80°C for
one hour under stirring conditions, and then was cooled down.
The crystals formed were collected, dissolved in a solution of
methanol and chloroform under heating conditions, and then
~~filtrated~~filtered. The obtained filtrate was distilled to remove
chloroform and cooled ~~down~~ to obtain 3.4 g of an orangish yellow
crystal of the styryl dye represented by Chemical Formula 40.
When measured in a conventional manner, the melting point of the
crystal was 233°C.

Page 45, please amend paragraph 1 as follows:

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Ten milliliters of dimethylformamide and 1 ml of iodomethyl were placed in a reaction vessel, and mixed with 1 g of the styryl dye obtained in Example 3. The resulting mixture was reacted at 100°C for one hour under stirring conditions, and then was cooled ~~down~~. The crystals formed were collected and recrystallized in ethanol to obtain 0.8 g of an orangish red crystal of the styryl dye represented by Chemical Formula 36. When measured in a conventional manner, the melting point of the crystal was 222-223°C.

Page 45, please amend paragraph 2 as follows:

Twelve milliliters of ethanol and 0.24 ml of piperidine were placed in a reaction vessel, and mixed with 3 g of 1,3-diethyl-2-methyl-5,6-dichlorobenzimidazolium = tosylate and 1.1 g of 4-nitrobenzaldehyde. The resulting mixture was reacted at 80°C for one hour under stirring conditions, and then was cooled ~~down~~. The crystals formed were collected, dissolved in a solution of methanol and water under heating conditions, and then ~~filtrated~~filtered. The obtained filtrate was distilled to concentrate ~~to a half volume of the solution~~ to half volume and cooled ~~down~~ to obtain 1.7 g of a yellow crystal of the styryl dye represented by Chemical Formula 23. When measured in a conventional manner, the melting point of the crystal was 287-290°C.

Page 47, please amend paragraph 1 as follows:

As shown in the visible absorption spectra of Fig. 3, the conventional related compound represented by Chemical Formula 61 has absorption maxima at a wavelength longer than 400 nm when formed in a solution and in a thin layer. ~~While, as~~ As shown in the results of Figs. 1 and 2 and Table 1, most of the styryl dyes of the present invention have absorption maxima at a wavelength of 400 nm or less when formed in a solution and in a thin layer, and the absorption ends to the side of longer-wavelength area extended to around 450 nm when formed in a thin layer. These results showed that the styryl dyes of the present invention were different from conventional related compounds and that they were sensitive to a laser beam with a wavelength of 450 nm or less, particularly, a laser beam with a wavelength around 350-450 nm, and substantially absorb such a laser beam.

Page 47, please amend paragraph 2 as follows:

Fifteen milligrams (mg) of either of the styryl dyes of the present invention represented by Chemical Formulae 9, 19, 23, 34, 36, 38, 40, and 44 was added to three milliliters (ml) of TFP. To the mixture was added 2 mg of nitrosodiphenylamine represented by Chemical Formula 62 which is used in ~~the~~

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conventional optical recording media as a light-resistant improver, followed by 5-min ultrasonic energization at ambient temperature to dissolve the contents in the solvent. Thereafter, in a conventional manner, a prescribed amount of the resulting solution was dropped on either surface of a polished glass substrate, 5 cm × 5 cm, while the glass substrate was rotated at a rotation rate of 1,200 rpm for one minute to uniformly coat the solution thereupon, and sequentially blown with hot air and cold air to form a thin layer on the glass substrate.

Page 48, please amend paragraph 1 as follows:

The resulting glass substrate coated with either of the styryl dyes was measured for transmittance (T_0) at a wavelength of 400 nm, and then set to the position seven cm away from a 500 W xenon lamp and exposed ~~with~~ to the light of the lamp for 25 min while sending a cold air to the substrate. Immediately after that, the resulting substrate was remeasured for transmittance (T) at a wavelength of 400 nm, and the transmittance of T and T_0 for each styryl dye was substituted for the Equation 1 to calculate the residual percentage (%) of the styryl dye. In parallel, control systems with no light-resistant improver for each styryl dye were provided.

Page 51, please amend paragraph 1 as follows:

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As shown in the results of Table 2, in the system with no light-resistant improvers, maximum 70% of the styryl dyes had changed with only 25_-min exposure of light to be incapable of exerting their inherent light absorbent properties. ~~While,~~ ~~when~~When used in combination with the nitroso compound represented by Chemical Formula 62, over 76 % of the styryl dyes still remained intact without being changed by the exposure. These results indicate that light-resistant improvers are useful to suppress undesirable changes of the styryl dyes of the present invention by exposure to natural- and artificial_--~~lights~~ light.

Page 51, please amend paragraph 2 as follows:

Either of the styryl dyes of the present invention represented by Chemical Formulae 8, 9, 19, 23, 34, 36, 38, 40, and 44 as a light absorbent was admixed with TFP to give a concentration of 3.0%(w/w), and the mixture was further added the nitroso compound represented by Chemical Formula 62 to give a concentration of 0.35%(w/w), heated for a while, followed by ultrasonically dissolving the contents. The resulting solution was in a conventional manner ~~filtrated~~ byfiltered through a membrane, coated in a rotatory manner over one side of an ~~aeryl-acrylic~~ disc substrate, 12 cm in diameter, to which had been transferred concaves for expressing synchronizing signals and addresses of tracks and

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sectors by an injection molding, to give a thickness of 200 nm. Thereafter, the substrate was spattered with silver to form a reflection layer, 100 nm in thickness, to be closely attached on the surface of the recording layer by vaporization, and the reflection layer was homogeneously coated in a rotatory manner with "DAICURE CLEAR SD1700", as a known ultraviolet ray hardening resin commercialized by Dainippon Ink and Chemicals, Inc., Tokyo, Japan, and irradiated to form a protection layer to be closely attached on the surface of the reflection layer, followed by obtaining nine types of optical recording media.

Page 52, please amend paragraph 1 as follows:

All of the optical recording media of this Example can be used for writing ~~written~~ large amounts of information in the form of documents, images, and ~~voices~~ sound at a relatively-high density by using a laser element with an oscillation wavelength of 450 nm or less.

Page 52, please amend paragraph 2 as follows:

As described above, the present invention was made based on the creation of novel styryl dyes and the findings of their industrially usable characteristics. Since the styryl dyes of the present invention have absorption maxima at a wavelength

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of 400 nm or less and substantially absorb a visible light with a wavelength around 400 nm, they have diversified uses in the fields of optical recording media, photochemical polymerization, dye laser, solar batteries, lithography, dyeing, etc., which need organic compounds with such properties. Particularly, among the styryl dyes, they, ~~which are being~~ sensitive to a laser beam with a wavelength of 450 nm or less, are advantageously useful as a material for a recording layer of high-density optical recording media such as DVD-Rs.

Page 52, please amend paragraph 3 as follows:

As compared with the conventional DVD-Rs which use polymethine dyes as a light absorbent and write information by using a laser beam with a wavelength of 635 nm or 650 nm, the optical recording media of the present invention, which include the styryl dyes of the present invention and use a laser beam with a wavelength of 450 nm or less as a writing light, can form minute pits at a relatively-narrower track pitch and a relatively-high density. Accordingly, when used the optical recording media of the ~~resent-present~~ invention, they can record a vast amount of information of characters, images, and ~~voices~~ sound at a relatively-high density, resulting in greatly lowering the cost of recording information per bit and recording

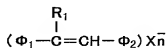
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animations for a long time, which need much recording volume, of course standing picture as a merit.

IN THE CLAIMS

1. A styryl dye represented by Formula 1:

Formula 1:

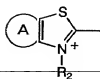


wherein in Formula 1, Φ_1 represents a heterocycle represented by any one of Formulae 2 to 8; Φ_2 represents an optionally substituted aromatic ring or heterocycle; R_1 represents a hydrogen atom, an aliphatic hydrocarbon group, ether group, acyl group, halogen, or cyano group, and the aliphatic hydrocarbon group, ether group, and acyl group may have a substituent; X^- represents ~~an arbitrary~~ counter ion; and "n" is a number of X^- ~~for keeping the~~ balance ~~of an~~ electric charge in the styryl dye:

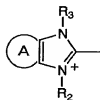
Formula 2:



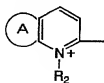
Formula 3:



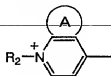
Formula 4:



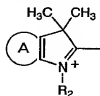
Formula 5:



Formula 6:



Formula 7:



Formula 8:



throughout Formulae 2 to 7, A represents an optionally substituted monocyclic- or polycyclic-aromatic ring or

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heterocycle; when A ~~does-is~~ not ~~exist-present~~ in Formulae 2 to 7, ~~a substituentone or more substituents~~ similar to those that are bound that combines for A may position to where may be in the position where A may exist is located; throughout Formulae 2 to 8, R₂ represents an optionally substituted aliphatic hydrocarbon group and R₃ represents a hydrogen or an optionally substituted aliphatic hydrocarbon group which is identical to or different from R₂.

2. (Amended) The styryl dye of claim 1, wherein said ϕ_2 is a nitrogen-atom-containing heterocycle which forms an ammonium salt.

3. (Amended) The styryl dye of claim 1 ~~or-2~~, wherein said counter ion is an organic metal complex anion with a light-resistant improving ability.

4. (Amended) The styryl dye of claim 1, ~~2-or-3~~, which has an absorption maximum at a wavelength of 400 nm or less.

5. (Amended) The styryl dye of claim 1, ~~2, 3-or-4~~, which substantially absorbs a visible light with a wavelength of 450 nm or less when formed in a thin layer.

6. (Amended) A light absorbent comprising the styryl dye of ~~any one of claims 1 to 5~~claim 1.

7. (Amended) The light absorbent of claim 6, which further contains one or more other organic dye compounds ~~that are~~ sensitive to a visible light.

8. (Amended) The light absorbent of claim 6 ~~or 7~~, which further contains one or more appropriate light-resistant improvers.

9. (Amended) The light absorbent of claim 6, ~~7 or 8~~ which is sensitive to a visible light with a wavelength of 450 nm or less when formed in a thin layer.

10. (Amended) An optical recording medium comprising the styryl dye of ~~any one of claims 1 to 5~~claim 1.

11. (Amended) The optical recording medium of claim 10, which further contains one or more other organic dye compounds ~~that are~~ sensitive to a visible light.

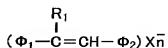
12. (Amended) The optical recording medium of claim

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10-~~or 11~~, which further contains one or more appropriate light-resistant improvers in a recording layer.

13. (Amended) The optical recording medium of claim 10, ~~11 or 12~~, which uses a laser beam with a wavelength of 450 nm or less as a writing light.

14. (Amended) A process for producing the styryl dye of Formula 1: A process for producing the styryl dye of any one of claims 1 to 5, comprising a step of reacting a compound, represented by Formula 9 having Φ_1 and R_1 corresponding to Formula 1, with a compound represented by Formula 10 having Φ_2 corresponding to Formula 1.



wherein in Formula 1, Φ_1 represents a heterocycle represented by any one of Formulae 2 to 8; Φ_2 represents an optionally substituted aromatic ring or heterocycle; R_1 represents a hydrogen atom, an aliphatic hydrocarbon group, ether group, acyl group, halogen, or cyano group, and the aliphatic hydrocarbon group, ether group, and acyl group may have a substituent; X^- represents a counter ion; and "n" is a number of X^- to balance the electric charge in the styryl dye;

In re Appl. KASADA4

Formula



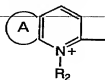
Formula 3:



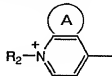
Formula 4:



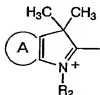
Formula 5:



Formula 6:



Formula 7:



Formula 8:

In re Appl. KASADA4



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102880.1:208660

throughout Formulae 2 to 7, A represents an optionally substituted monocyclic or polycyclic-aromatic ring or heterocycle; when A is not present in Formulae 2 to 7, a substituent similar to A is in the position filled by A; throughout Formulae 2 to 8, R₂ represents an optionally substituted aliphatic hydrocarbon group and R₃ represents a hydrogen or an optionally substituted aliphatic hydrocarbon group which is identical to or different from R₂.
comprising reacting a compound of Formula 9

Formula 9:



with a compound of Formula 10

Formula 10:



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DESCRIPTION

STYRYL DYES

5

Technical Field

The present invention relates to organic dye compounds, and more particularly, to styryl dyes which have absorption maxima at a wavelength of 400 nm or less.

10

Background Art

In a multimedia age, optical recording media such as CD-R (a write-once memory using compact disc) and DVD-R (a write-once memory using digital video disc) are now greatly highlighted. Most of the conventional optical recording media can be roughly classified into inorganic optical recording media which have recording layers composed of inorganic substances such as tellurium, selenium, rhodium, carbon, or carbon sulfide; and organic optical recording media which have recording layers mainly composed of light absorbers containing organic dye compounds.

Among these optical recording media, organic optical recording media can be usually prepared by dissolving a polymethine dye in an organic solvent such as 2,2,3,3-tetrafluoro-1-propanol (abbreviated as "TFP" hereinafter), spun coating the solution on the surface of a polycarbonate substrate, drying the solution to form a recording layer, and sequentially forming and coating onto

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the surface of the recording layer a reflection layer comprising a metal such as gold, silver or copper, and a protection layer comprising an ultraviolet ray hardening resin. When compared with inorganic optical recording media, organic ones may have the drawback that their recording layers are susceptible to change under environmental lights such as reading- and natural-lights. Organic optical recording media, however, have the merit that they can be made into optical recording media at a lesser cost because their recording layers can be directly formed by coating light absorbents in solution on the surface of substrates. Further, organic optical recording media composed of organic materials are now mainly used as low-cost optical recording media because they are substantially free of corrosion even when contacted with moisture or sea water and because information recorded therein in a prescribed format can be read out by using commercially available read-only readers after the establishment of thermal-deformation-type optical recording media, a kind of organic optical recording media.

What is urgently required in organic optical recording media is to increase their storage capacity to suit to this multimedia age. The research for such an increment, which is now being eagerly continued in this field, is to increase a recording capacity per one side from 4.7 GB (giga bite) to 15 GB or more by shortening the wavelength for writing information to 450 nm or less from 635-650 nm which are commonly used. The high-density optical recording media have the capacity to record movie and animations for six hours in picture quality of standard television or for two hours even in relatively-high picture quality of high definition television.

However, since most of organic dye compounds now used in the conventional optical recording media do not suit to a visible light with a wavelength of 450 nm or less, they could not fulfil the need for high-storage density required in many fields.

5 In view of the foregoing, the object of the present invention is to provide organic dye compounds which have absorption maxima at a wavelength of 400 nm or less and substantially absorb a visible light with a wavelength of 450 nm or less when formed in a thin layer, and to provide uses thereof.

10 Summary of Invention

The present inventors eagerly studied and screened compounds. As a result, they found that specific styryl dyes (may
15 be called "styryl dyes" hereinafter), which are obtainable through a step of reacting a quaternary ammonium salt of nitrogen heterocyclic compound having an active methyl- or active methylene-group with an aldehyde compound, have absorption maxima at a wavelength of 400 nm or less and substantially absorb a visible light
20 with a wavelength around 400 nm. The present inventors confirmed that the styryl dyes, which substantially absorb a visible light with a wavelength of 450 nm or less when formed in a thin layer, form remarkably-minute pits at a relatively-high density in optical recording media by irradiating with a laser beam with a wavelength
25 of 450 nm or less. The present invention was made based on the creation of novel styryl dye and the discovery of their industrially useful characteristics.

Brief Description of Drawings

FIG. 1 is visible absorption spectra of one of the styryl dyes of the present invention when in a solution form and in a thin layer form, respectively.

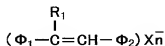
FIG. 2 is visible absorption spectra of another styryl dye of the present invention when in a solution form and in a thin layer form, respectively.

FIG. 3 is visible absorption spectra of a conventional related compound when in a solution form and in a thin layer form, respectively.

Best Mode for carrying out the Invention

The present invention relates to the styryl dyes represented by Formula 1.

Formula 1:



In Formula 1, R_1 represents a hydrogen atom, an aliphatic hydrocarbon group, ether group, acyl group, halogen, or cyano group. The aliphatic hydrocarbon group in R_1 represented that having up to six carbon atoms, usually, from one to five carbon atoms such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, *sec*-butyl, *tert*-butyl, pentyl, isopentyl, neopentyl, or *tert*-pentyl group. Examples of the ether groups are methoxy, ethoxy, propoxy,

isopropoxy, buthoxy, *tert*-buthoxy, pentoxy, and phenoxy groups. Examples of the acyl groups are formyl, acetyl, propionyl, butyryl, *tert*-butyryl, benzoyl, *o*-tolyl, *m*-tolyl, and *p*-tolyl groups. Examples of the halogens are fluorine, chlorine, bromine, and iodine.

- 5 One or more of the hydrogens in the aliphatic hydrocarbon group, ether group, and acyl group may be replaced with a short-chain aliphatic hydrocarbon group such as methyl, ethyl, or propyl group; a short-chain ether group such as methoxy, ethoxy, or propoxy group; or a halogen such as fluorine, chlorine, bromine, or iodine.

- 10 In Formula 1, ϕ_1 represents a heterocycle which is represented by any one of Formulae 2 to 8.

Formula 2:



Formula 3:

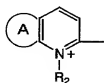


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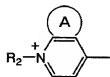
Formula 4:



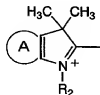
Formula 5:



Formula 6:



5 Formula 7:



Formula 8:



Throughout Formulae 2 to 7, A represents a monocyclic-
10 or polycyclic-aromatic-ring or heterocycle. Examples of the
aromatic ring are benzene- and naphthalene-rings. Examples of the
heterocycle are pyridine, quinoline, and quinoxaline rings. The
aromatic ring and heterocycle may have one or more substituents,
for example, a halogen atom such as fluorine, chlorine, bromine,
15 or iodine atom; a short-chain aliphatic hydrocarbon group such as
methyl, trifluoromethyl, ethyl, propyl, isopropyl, butyl, isobutyl,
sec-butyl, or *tert*-butyl group; a short-chain ether group such as

methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, buthoxy, or *tert*-buthoxy group; an alkylaminosulfonyl group such as methylaminosulfonyl, dimethylaminosulfonyl, ethylaminosulfonyl, diethylaminosulfonyl, propylaminosulfonyl, dipropylaminosulfonyl, 5 butylaminosulfonyl, or dibutylaminosulfonyl group; an aromatic hydrocarbon group such as phenyl, biphenyl, *o*-tolyl, *m*-tolyl, *p*-tolyl, xyl, mesityl, *o*-cumenyl, *m*-cumenyl, *p*-cumenyl, or naphthyl group; an ester group such as methoxycarbonyl, trifluoromethoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, 10 acetox, trifluoroacetox, or benzoyloxy group; nitro group, cyano group, carboxy group, or sulfo group. When A does not exist in Formulae 2 to 7, one or more substituents similar to that combines for A may position to where A may exist.

Throughout Formulae 2 to 8, R₂ represents an aliphatic 15 hydrocarbon group selected from those having up to eight carbon atoms, usually, up to six carbon atoms such as methyl, ethyl, propyl, isopropyl, isopropenyl, 1-propenyl, 2-propenyl, 2-propynyl, butyl, isobutyl, *sec*-butyl, *tert*-butyl, 2-butenyl, 2-buty, 1,3-butadienyl, pentyl, isopentyl, neopentyl, 1-methylpentyl, 2- 20 methylpentyl, 2-pentenyl, 2-pentynyl, 2-penten-4-ynyl, hexyl, or isohexyl group. The aliphatic hydrocarbon group may have one or more substituents, for example, a halogen such as fluorine, chlorine, bromine, or iodine; an ether group such as methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, buthoxy, *tert*-buthoxy, benzyloxy, or 25 phenoxy group; an ester group such as methoxycarbonyl, trifluoromethoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, acetox, trifluoroacetox, or benzoyloxy group; a carboxy group;

a sulfo group; a hydroxy group; an amido group such as acetylamino or benzoylamino group; an aromatic hydrocarbon group such as phenyl, biphenyl, *o*-tolyl, *m*-tolyl, *p*-tolyl, xyl, mesityl, *o*-cumenyl, *m*-cumenyl, *p*-cumenyl, or naphthyl group; or heterocyclic group such as 2-pyridyl, 2-quinolyl, 2-tetrahydropyranyl, 2,2-dimethyl-1,3-dioxolane-4-yl, 1,3-dioxolane-2-yl, 3,5-dimethylisooxazole-4-yl, 3-piperidinyl, piperidino, morpholino, 1-piperazinyl, pyrrolidine-1-yl, 1-methyl-2-pyrrolidinyl, 2-benzimidazolyl, 5-uracil, or benzotriazole-1-yl group.

In Formula 4, R₁ represents a hydrogen atom or an aliphatic hydrocarbon group which is identical to or different from R₂. Representative example of the aliphatic hydrocarbon group is short-chain those such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, *sec*-butyl, or *tert*-butyl group, which may have one or more substituents, for example, an ether group such as methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, butoxy, or *tert*-butoxy group; an ester group such as methoxycarbonyl, trifluoromethoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, acetoxyl, trifluoroacetoxyl, or benzoyloxy group; or an aromatic hydrocarbon group such as phenyl, *o*-tolyl, *m*-tolyl, *p*-tolyl, xylyl, mesityl, *o*-cumenyl, *m*-cumenyl, *p*-cumenyl, or nitrophenyl group.

Furthermore, in Formula 1, ϕ_2 represents a monocyclic or polycyclic-aromatic ring or heterocycle which may include one or more nitrogen atoms into the ring, for example, benzene, pyridine, pyrimidine, pyridazine, naphthalene, quinoline, or quinoxaline ring. The aromatic ring and heterocycle may have one or more substituents. The substituents are selected from those having up to eight carbon

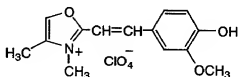
atoms, usually, one to six carbon atoms, for example, an aliphatic hydrocarbon group such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, *sec*-butyl, *tert*-butyl, pentyl, isopentyl, neopentyl, *tert*-pentyl, 1-methylpentyl, 2-methylpentyl, hexyl, or isohexyl group; a short-chain ether group such as methoxy, trifluoromethoxy, ethoxy, propoxy, isopropoxy, butoxy, or *tert*-butoxy group; an amido group such as acetylamino or benzoylamino group; a halogen such as fluorine, chlorine, bromine, or iodine; and cyano group, nitro group, hydroxy group, sulfo group, or carboxy group. When ϕ_2 is a nitrogen-atom-containing heterocycle, the nitrogen atom may combine with the similar aliphatic hydrocarbon group as in R_2 in ϕ to form an ammonium salt. When ϕ_2 has two hydroxy groups as a substituent and the hydroxy groups combine with adjacent carbon atoms in ϕ_2 , the hydroxy groups may react with an oxo compound such as formalin to form a cyclic structure such as a dioxole ring.

X^- in Formula 1 represents an arbitrary counter ion and is usually selected from inorganic acid ions such as fluoric, chrolic, bromic, iodic, perchrolic acid, periodic acid, phosphoric acid hexafluoride, antimony hexafluoride, tin acid hexafluoride, phosphoric acid, fluoroboric acid, and tetrafluoroboric acid ions; organic acid ions such as thiocyanic acid, benzensulfonic acid, naphthalenesulfonic acid, naphthalenedisulfonic acid, *p*-toluenesulfonic acid, alkylsulfonic acid, benzencarboxylic acid, alkylcarboxylic acid, trihaloalkylcarboxylic acid, alkylsulfonic acid, trihaloalkylsulfonic acid, nicotinic acid, and tetracyanoquinonedimethane ions; and organic metal complex ions such as azo, bisphenyldithiol, thiocatechol chelate,

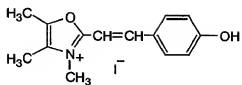
thiobisphenolate chelate, and bisdiol- α -diketone. "n" is a number of X⁻ to keep a balance of the electric charge in styryl dyes, and is usually an integer selected from one and two.

The present invention relates to styryl dyes having the structures as mentioned above, particularly, to those which have absorption maxima at a wavelength of 400 nm or less, and desirably those which substantially absorb a visible light with a wavelength of 450 nm or less when formed in a thin layer. Concrete examples of the styryl dyes are those represented by Chemical Formulae 1 to 60. Since all of them have absorption maxima at a wavelength of 400 nm or less and substantially absorb a visible light with a wavelength around 400 nm, they have diversified uses in the fields, which need organic compounds that absorb the aforesaid visible light. Among the styryl dyes, those, which substantially absorb a visible light with a wavelength of 450 nm or less when formed in a thin layer, particularly, absorb the visible light in the absorption end of longer wavelength region of the absorption maximum, are remarkably useful as a material for a recording layer of high-density-optical recording media such as DVD-Rs for coming generation.

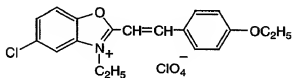
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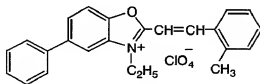
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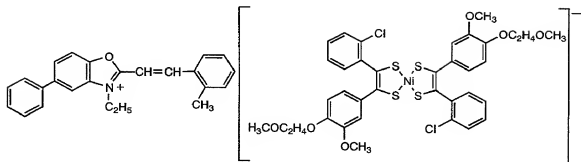
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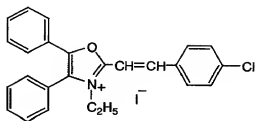
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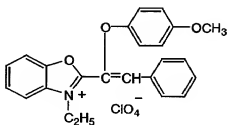
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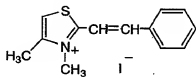
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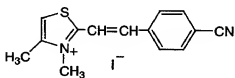
Chemical Formula 7:



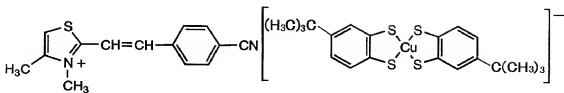
5 Chemical Formula 8:



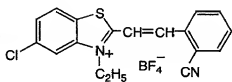
Chemical Formula 9:



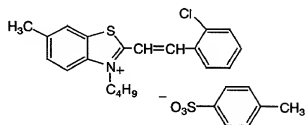
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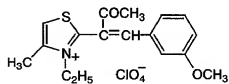
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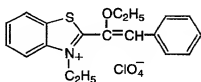
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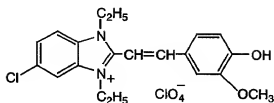
5 Chemical Formula 13:



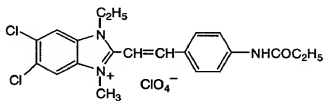
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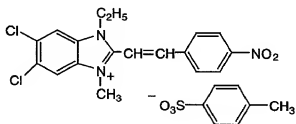
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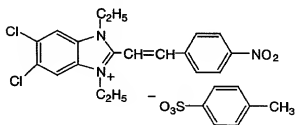
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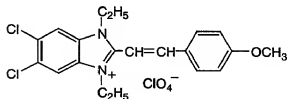
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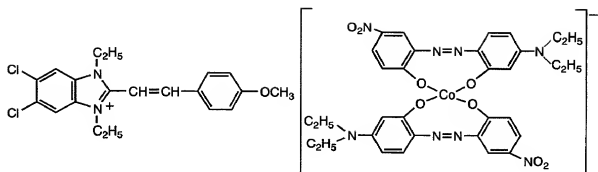
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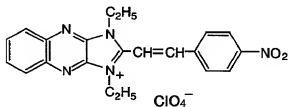
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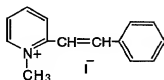
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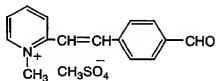
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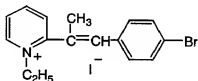
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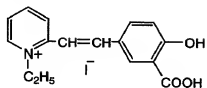
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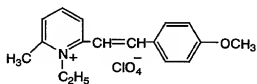
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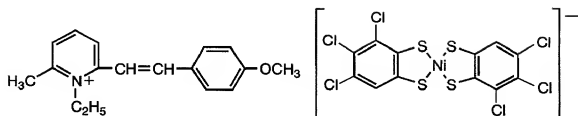
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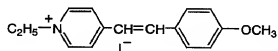
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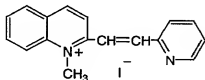
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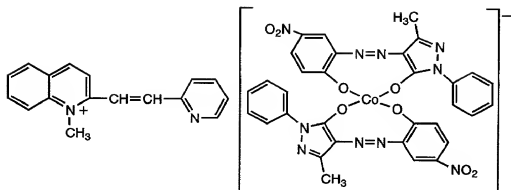
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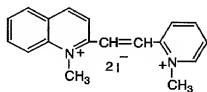
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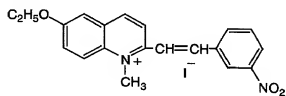
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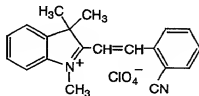
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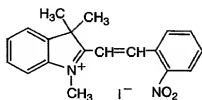
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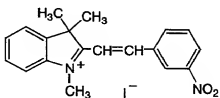
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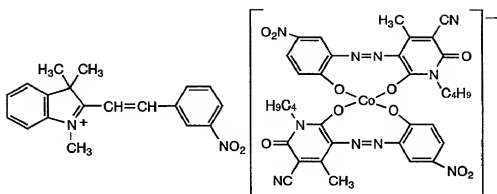
Chemical Formula 39:



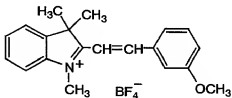
Chemical Formula 40:



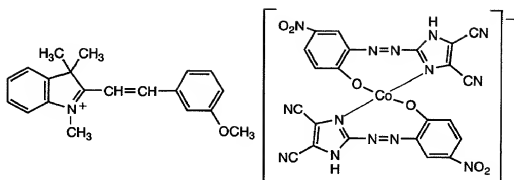
5 Chemical Formula 41:



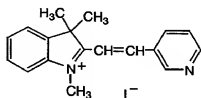
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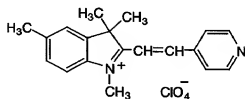
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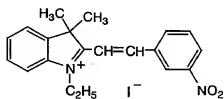
Chemical Formula 44:



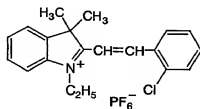
5 Chemical Formula 45:



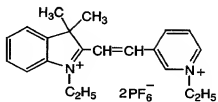
Chemical Formula 46:



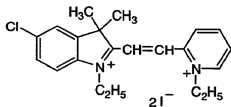
Chemical Formula 47:



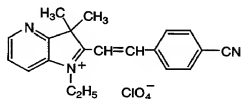
Chemical Formula 48:



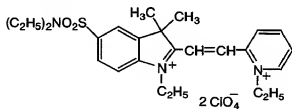
Chemical Formula 49:



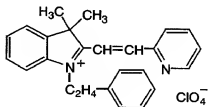
5 Chemical Formula 50:



Chemical Formula 51:

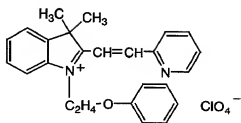


Chemical Formula 52:

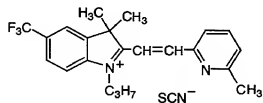


10

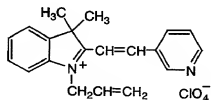
Chemical Formula 53:



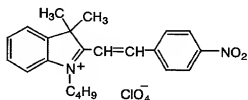
Chemical Formula 54:



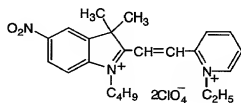
5 Chemical Formula 55:



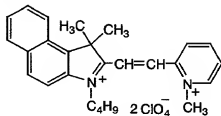
Chemical Formula 56:



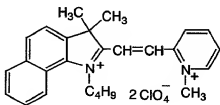
Chemical Formula 57:



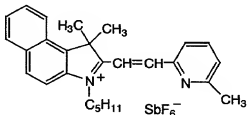
Chemical Formula 58:



Chemical Formula 59:



5 Chemical Formula 60:



The styryl dyes of the present invention can be prepared by various manners. They can be preferably produced through a step of reacting a quaternary ammonium salt of nitrogen-atom-containing heterocyclic compound having either an active methyl- or active methylene-group with an aldehyde compound from an economical view point. With the manner, the styryl dyes of the present invention can be produced in a desired yield by reacting a compound represented by Formula 9 having ϕ_1 and R₁ corresponding to those in Formula 1 with a compound represented by Formula 10 having ϕ_2 corresponding to those in Formula 1.

Formula 9:



Formula 10:



5 For example, appropriate amounts (usually about equal mols)
of compounds represented by Formulae 9 and 10 are placed in a reaction
vessel, and the resulting mixture is dissolved in an adequate solvent,
if necessary, and then reacted at ambient temperature or over under
heating and stirring conditions, for example, a reflux in the
10 presence of an appropriate amount of a basic compound such as sodium
hydroxide, potassium hydroxide, sodium carbonate, potassium
carbonate, sodium hydrogencarbonate, potassium hydrogencarbonate,
ammonia, triethylamine, piperidine, pyridine, pyrrolidine, aniline,
N,N-dimethylaniline, N,N-diethylaniline, or N-methyl-2-
15 pyrrolidone; an acidic compound such as hydrochloric acid, sulfuric
acid, nitric acid, methanesulforic acid, p-toluenesulfonic acid,
acetic acid, acetic anhydride, propionic anhydride, trifluoroacetic
acid, or trifluorosulfonic acid; or a Lewis acidic compound such
as aluminium chloride, zinc chloride, tin tetrachloride, or titanium
20 tetrachloride.

As to solvents which can be used in the present invention,
for example, hydrocarbons such as pentane, hexane, cyclohexane,
octane, benzene, toluene, and xylene; halides such as carbon

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tetrachloride, chloroform, 1,2-dichloroethane, 1,2-dibromoethane, trichloroethylene, tetrachloroethylene, chlorobenzene, bromobenzene, and α -dichlorobenzene; alcohols and phenols such as methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, 5 isobutyl alcohol, isopentyl alcohol, cyclohexanol, ethylene glycol, propylene glycol, 2-methoxyethanol, 2-ethoxyethanol, phenol, benzyl alcohol, cresol, diethylene glycol, triethylene glycol, and glycerin; ethers such as diethyl ether, diisopropyl ether, tetrahydrofuran, tetrahydropyran, 1,4-dioxane, anisole, 1,2-10 dimethoxyethane, diethylene glycol dimethyl ether, dicyclohexyl-18-crown-6, methylcarbitol, and ethylcarbitol; ketones such as furfural, acetone, ethyl methyl ketone, and cyclohexanone; acids and acidic derivatives such as acetic acid, acetic anhydride, trichloroacetic acid, trifluoroacetic acid, 15 propionic anhydride, ethyl acetate, butyl carbonate, ethylene carbonate, propylene carbonate, formamide, N-methylformamide, N,N-dimethylformamide, N-methylacetoamide, N,N-dimethylacetoamide, hexamethylphosphoric triamide, and phosphoric trimethyl; nitriles such as acetonitrile, propionitrile, 20 succinonitrile, and benzonitrile; nitro compounds such as nitromethane and nitrobenzene; sulfur-containing compounds such as dimethylsulfoxide and sulfolane; and water can be used and arbitrarily used in a combination, if necessary.

In general, when solvents are used, the greater the volume 25 of solvents the lower the reaction efficiency, on the contrary, the lower the volume of solvents the more difficult, homogeneous heating and stirring of the contents becomes and undesirable side reaction

may easily occur. Thus, the volume of solvents is desirably set to a level up to 100-folds, usually, in the range of 5-50-folds to the total volume of material compounds by weight. The reaction completes within 10 hours, usually, 0.5-10 hours, depending on the kinds of material compounds and the reaction conditions. The reaction progress can be monitored by conventional methods, for example, thin layer chromatography, gas chromatography, and high-performance liquid chromatography. After completion of the reaction, the styryl dyes of the present invention with desired counter ions are obtainable by providing the reaction mixture to a conventional counter ion-exchange reaction, if necessary. Thus, all of the styryl dyes represented by Chemical Formulae 1 to 60 are obtainable in a desired yield by the above method. Compounds represented by Formulae 9 and 10 are obtainable in accordance with conventional methods to form a cyclic nuclear in styryl dyes.

The styryl dyes thus obtained can be used in the form of a reaction mixture without any further treatment, but usually used after purified by conventional manners, which are generally used for purifying their related compounds, such as dissolution, extraction, separation, decantation, filtration, concentration, thin layer chromatography, column chromatography, gas chromatography, high-performance liquid chromatography, distillation, crystallization, and sublimation. If necessary, these methods can be used in combination. In using in dye lasers and optical recording media such as DVD-Rs, it is preferable that the styryl dyes of the present invention are distilled, crystallized, and/or sublimated prior to use.

The light absorbents of the present invention comprise one or more of the styryl dyes and include those which utilize the nature of the styryl dyes which substantially absorb visible light with a wavelength around 400 nm. The compositions or the physical forms of the light absorbents are not restricted. Therefore, the light absorbents of the present invention may comprise one or more other elements in addition to the styryl dyes, depending on use. One of the fields, which the light absorbents of the present invention can be advantageously used, is of optical recording media. In this field, the light absorbents of the present invention are suitable as a material for recording layers of optical recording media using a laser beam with a wavelength of 450 nm or less as a writing light. In using the light absorbents of the present invention in optical recording media, unless the use deviates from the object of the present invention, the styryl dyes can be mixed with other light absorbents comprising other organic dye compounds which are sensitive to visible light and one or more conventional materials used in optical recording media, such as a light-resistant improver, binder, dispersing agent, flame retardant, lubricant, antistatic agent, surfactant, thermal interference inhibitor, plasticizer, color fixing agent, developer, and dissolving assistant, if necessary.

Explaining the uses of the light absorbents of the present invention by exemplifying organic optical recording media (organic heat-deformative-type optical recording media), the optical recording media of the present invention can be prepared in accordance with conventional organic recording media because the

styryl dyes of the present invention require no special treatment and step which are commonly used in this art. For example, one or more of the styryl dyes of the present invention can be mixed with one or more other organic dye compounds which are sensitive to visible light, and one or more conventional light-resistant improvers, binders, dispersing agents, flame retardants, lubricants, antistatic agents, surfactants, thermal interference inhibitor, and plasticizers to control the reflection rate and the light absorption rate in a recording layer, if necessary. The resulting mixture is dissolved in an organic solvent, and the solution is uniformly coated over either surface of substrates by means of spraying, soaking, roller coating, or rotatory coating method; and dried to form a recording layer, a thin layer comprising light absorbents, and if necessary, followed by forming a reflection layer to be closely attached onto the recording layer by the method of vacuum deposition, chemical vapor deposition, sputtering, or ion plating using metals such as gold, silver, copper, platinum, aluminium, cobalt, tin, nickel, iron, and chromium or using conventional organic materials for reflection layers to impart a reflection efficiency which the written information can be read, for example, 20% or more, and preferably 30% or more; and spun coating over the recording layer ultraviolet-ray-hardening resins or thermosetting resins, which contain flame retardants, stabilizers, and/or antistatic agents, and then hardening the coated resins by either irradiating light or heating to form a protection layer to be closely attached onto the reflection layer to protect the recording layer from scratches, dusts, spoils, etc. Thereafter, if necessary, a pair of the above

substrates with recording-, reflection-, and recording-layers are
faced and attached together using, for example, adhesives or viscous
sheets; or protection plates, which are comprising the same
materials and shapes as the substrates, are attached to the
5 protection layers of the substrates.

As another organic dye compounds usable in combination with
the styryl dyes of the present invention, dye compounds are
particularly restricted as long as they are sensitive to visible
light and can modulate the light reflection- and the light
10 absorption-rates of recording layers of optical recording media.
As the organic dye compounds, the following compounds can be used
in an arbitrary combination, if necessary: Acridine dye,
azaannulene dye, azo dye, azo metal complex dye, anthraquinone dye,
indigo dye, indanthrene dye, oxazine dye, xanthene dye, dioxazine
15 dye, thiazine dye, thioindigo dye, tetrapyrapolphyradine dye,
triphenylmethine dye, triphenylthiazine dye, naphthoquinone dye,
pyrromethene dye, phthalocyanine dye, benzoquinone dye, benzopyran
dye, benzofuranone dye, polyphyrin dye, rhodamine dye, and
polymethine dye such as cyanine dye, merocyanine dye, oxonol dye,
20 azulenium dye, squalilium dye, styryl dye, pyririum dye,
thiopyririum dye, and phenanthrene dye which same or different rings
are bound to the both ends of polymethine chain such as monomethine,
dimethine, trimethine, tetramethine, pentamethine, hexamethine, or
heptamethine. The chain and rings can have one or more substituents.
25 Examples of the ring are imidazolin ring, imidazole ring,
benzoimidazole ring, α -naphthimidazole ring, β -naphthimidazole
ring, indole ring, isoindole ring, indolenine ring, isoindolenine

The light-resistant improvers are, for example, nitroso compounds such as nitrosodiphenylamine, nitrosoaniline, nitrosophenol, and nitrosonaphthol; and metal complexes such as dithiolate metal complexes, for example, "NKX-1199" (bis[2'-chloro-3-methoxy-4-(2-methoxyethoxy)dithiobenzyl] nickel) produced by Hayashibara Biochemical Laboratories, Inc., Okayama, Japan, and formazane metal complexes, which all can be arbitrarily used in combination, if necessary. Preferable light-resistance improvers are those which contain metal complexes, and most preferable ones are metal complexes, more preferably, formazane compounds which have a pyridine-ring at C-5 and a pyridine- or furan-ring at C-3 of formazane skeleton as disclosed in Japanese Patent Application No. 163,036/99, titled "Formazane metal complexes", and complexes to metals such as nickel, zinc, cobalt, iron, copper, and palladium, which have the tautomer of the aforesaid compounds as a ligand. In the case of using such a light-resistance improver in combination, the styryl dyes of the present invention can be effectively prevented from undesirable changing in deterioration, fading, color changing, and quality changing, which are inducible by environmental lights such reading- and natural-lights, without lowering the solubility of the light absorbents of the present invention in organic solvents and substantially deteriorating preferable optical features. As to a composition ratio, 0.01-5 moles of a light-resistance improver(s), desirably, 0.1-1 moles, can be incorporated into one mole of the styryl dye(s) while increasing or decreasing the ratio. The light-resistant improver(s) are not necessarily other compound(s)

which are separately exist from the styryl dyes of the present invention, and if necessary, they can be formulated into salts, complexes, or compounds by combining the styryl dyes with organic metal complex anions, which are capable of improving the light resistance as disclosed in Japanese Patent Kokai Nos. 19,355/89, 139,043/93, 323,478/97, and 6,651/98, such as conventional those of azo, bisphenyldithiol, phenyldithiol, thiocatecholchelate, thiobisphenolatechelate, or bisdithiol- α -diketone by using appropriate spacers and crosslinking agents such as alkoxides or cyarates of titanium, zirconium, aluminium, etc., or complexes of these metal elements having carbonyl compounds or hydroxy compounds as ligands.

The styryl dyes of the present invention have satisfactorily-high solubility in organic solvents without substantially causing undesirable problems in actual use, and do not particularly restrict the types of organic solvents used for coating the light absorbents of the present invention over substrates. Thus, in the preparation of optical recording media according to the present invention, arbitrary organic solvents can be selected from the following conventional ones which are arbitrarily used in combination, if necessary: TFP frequently used to prepare optical recording media and the following organic solvents other than TFP: For example, hydrocarbons such as benzene, toluene, and xylene; halides such as carbon tetrachloride, chloroform, 1,2-dichloroethane, 1,2-dibromoethane, trichloroethylene, tetrachloroethylene, chloroenezene, bromobenzene, and α -dichlorobenzene; alcohols and phenols such as

methanol, ethanol, propanol, isopropanol, 2,2,2-trifluoroethanol, butanol, isobutanol, isopentanol, cyclohexanol, ethylene glycol, propylene glycol, 2-methoxyethanol (methyl cellosolve), 2-ethoxyethanol (ethyl cellosolve), phenol, benzyl alcohol, cresol, diethylene glycol, triethylene glycol, glycerine, and diacetone alcohol; ethers such as diethyl ether, diisopropyl ether, tetrahydrofuran, tetrahydropyran, 1,4-dioxane, anisole, 1,2-dimethoxyethane, dicyclohexyl-18-crown-6, methylcarbitol, and ethylcarbitol; ketones such as furfural, acetone, 1,3-diacetylacetone, ethyl methyl ketone, and cyclohexanone; esters such as ethyl acetate, butyl acetate, ethylene carbonate, propylene carbonate, and trimethyl phosphate; amides such as formamide, N-methylformamide, N,N-dimethylformamide, and hexamethylphosphoric triamide; nitriles such as acetonitrile, propionitrile, and succinonitrile; nitro compounds such as nitromethane and nitrobenzene; amines such as ethylene diamine, pyridine, piperidine, morpholine, and N-methylpyrrolidone; and sulfur-containing compounds such as dimethylsulfoxide and sulfolane.

Particularly, since the styryl dyes of the present invention have relatively-high solubilities in easily volatile organic solvents such as TFP, methyl cellosolve, ethyl cellosolve, and diacetone alcohol, they are substantially free from dye crystallization when they are sequentially dissolved in the organic solvents, coated on substrates, and dried; and do not cause inconsistent thickness and surface of the formed recording layers. Most of the styryl dyes of the present invention exert desirable

solubilities in non-halogen solvents, for example, cellosolves such as methyl cellosolve and ethyl cellosolve, alcohols such as diacetone alcohol, and ketones such as ethyl methyl keton and cyclohexanone. When the styryl dyes of the present invention

5 dissolve in the non-halogen solvents and the obtained solutions are coated over the plastic substrates, the solvents neither damage the substrates nor pollute the environment as a merit.

Conventional substrates can be used in the present invention and usually processed with suitable materials, for example,

10 into discs, 12 cm in diameter and 0.1-1.2 mm in thickness, to suit to final use by the methods such as compression molding, injection molding, compression-injection molding, photopolymerization method (2P method), thermosetting integral method, and lightsetting integral method. These discs can be used singularly or plurally

15 after appropriately attached them together with adhesive sheets or adhesive agents, etc. In principal, any materials for substrates can be used in the present invention as long as they are substantially transparent and have light transmissivity of at least 80%, and preferably 90% or more at the wavelengths ranging from 350 nm to

20 800 nm. Examples of such materials are glasses, ceramics, and others such as plastics including polyacrylate, polymethyl methacrylate, polycarbonate, polystyrene (styrene copolymer), polymethylpenten, polyester, polyolefin, polyimide, polyetherimide, polysulfone, polyethersulfone, polyarylate, polycarbonate/polystyrene alloy,

25 polyestercarbonate, polyphthalatecarbonate, polycarbonateacrylate, non-crystalline polyolefin, methacrylate copolymer, diallylcarbonatediethylene-glycol, epoxy resin, and

phenol resin, where polycarbonate and acrylate are most frequently used. In plastic substrates, concaves for expression of synchronizing signals and addresses of tracks and sectors are usually transferred to the internal circle of the tracks during their formation. The present invention does not restrict the form of concaves and the concaves are preferably formed to give 0.3-0.8 μ m in average wide and 50-150 nm in depth.

The styryl dyes of the present invention can be prepared into 0.5-5%(w/w) solutions of the organic solvents as mentioned above while considering the viscosity of the solutions, and then uniformly coated over a substrate to form a recording layer with 10-1,000 nm, preferably, 50-300 nm in thickness. Prior to the coating, a preliminary layer can be formed over the substrate to improve the protection and the adhesion ability of the substrate, if necessary. Materials of the preliminary layer are, for example, high-molecular substances such as ionomer resins, polyamide resins, vinyl resins, natural resins, silicon, and liquid rubbers. In the case of using binders, the following polymers can be used alone or in combination in a weight ratio of 0.01-10 times of the light absorbent(s): Cellulose esters such as nitrocellulose, cellulose phosphate, cellulose sulfate, cellulose acetate, cellulose propionate, cellulose lactate, cellulose palmitate, and cellulose acetate/propionate; cellulose ethers such as methyl cellulose, ethyl cellulose, propyl cellulose, and butyl cellulose; vinyl resins such as polystyrene, poly(vinyl chloride), poly(vinyl acetate), poly(vinyl acetal), poly(vinyl butyral), poly(vinyl formal), poly(vinyl alcohol), and poly(vinyl pyrrolidone); copolymer resins

in reading recorded informations, although the power levels can be varied depending on the types and ratios of the light-resistant improvers used in combination with the light absorbents of the present invention and further the writing speed: Generally, the levels can be controlled to outputs of over 5 mW for writing, usually in the range of 10-50 mW, and to outputs of 5 mW or lower for reading, usually in the range of 0.1-5 mW. The recorded informations are read by detecting the changes of both the reflection light level and the transmission light level in the pits and the pit-less part on the surface of optical recording media by the light pick-up manner.

Accordingly, in the optical recording media of the present invention, remarkably-minute pits with a pit width of below $0.4 \mu\text{m}$ and a track pitch of below $0.74 \mu\text{m}$ used in the conventional DVD-R, can be formed at a relatively-high density by using a laser beam with an oscillating wavelength of 450 nm. For example, in using a substrate, 12 cm in diameter, it can realize an extremely-high density optical recording medium with an optical recording capacity far exceeding 4.7 GB (giga bytes) per one side, i.e., that of about two hours of information of sound and images in relatively-high image quality of high definition television which could hardly be attained in the commonly used DVD-Rs.

Since the optical recording media of the present invention can record information of characters, images, and sound at a relatively-high density, they are advantageously useful as recording media for professional and family uses to record and care for documents, data, and computer software. Particular examples of the types of industries and the forms of information to which

As a rather specific use, the optical recording media of the present invention can be used to edit and proofread compact discs, digital video discs, laser discs, MD (a mini disc as an information recording system using photomagnetic disc), CDV (a laser disc using compact disc), DAT (an information recording system using magnetic tape), CD-ROM (a read-only memory using compact disc), DVD-ROM (a read-only memory using digital video disc), DVD-RAM (a writable and readable memory using digital video disc), digital photos, movies, computer graphics, publishing products, broadcasting programs, commercial messages, computer programs, video software, audio software, game software, etc.; and used as external program recording means for large size of computers and car navigation systems.

Hereinbefore, the use of the light absorbents of the present invention in the field of optical recording media has been mainly explained with reference to their application examples to high-density optical recording media which use a laser beam with a wavelength of 450 nm or less as a writing light. However, in the field of optical recording media, the light absorbents of the present invention can be also advantageously used as materials for changing or regulating the optical absorption rate or the optical reflection rate in the existing optical recording media such as CD-Rs, DVD-Rs, and other high-density optical recording media by using in combination, for example, with one or more other organic dye compounds which are sensitive to a laser beam with a wavelength around 635-650 nm or 775-795 nm. When applied to organic-ablation type optical recording media using a laser beam with a wavelength of 450

nm or less as a writing light, the styryl dyes of the present invention can be used not to directly form pits on substrates but to indirectly form pits in such a manner that the excitation energy of a laser beam with a wavelength of 450 nm or less is allowed to transfer to the aforesaid organic dye compounds via the styryl dyes by using the styryl dyes along with one or more other organic dye compounds which are sensitive to a laser beam with a longer wavelength, e.g., a laser beam with a wavelength around 635-650 nm or 775-795 nm, resulting in a decomposition of the organic dye compounds. The optical recording media as referred to in the present invention mean optical recording media in general which use the characteristics of the specific styryl dyes of the present invention that have the absorption maxima at a wavelength of 400 nm or less and substantially absorb a visible light with a wavelength around 400 nm in addition to organic thermal-deformed optical recording media, thermal coloration method using the chemical reaction of coloring agents and developers using the heat generated when organic dye compounds absorb light, and the technique called "moth-eye type technique" which uses the phenomenon of that the above heat smoothes the pattern of periodical unevenness provided on the surface of the substrates.

Since the styryl dyes of the present invention have absorption maxima at a wavelength of 400 nm or less and substantially absorb a visible light with a wavelength around 400 nm, the light absorbents of the present invention containing the styryl dyes can be advantageously useful to the aforesaid optical recording media and also used as materials for polymerizing polymerizable compounds by exposure to visible light, sensitizing solar batteries, light

absorptive materials for lithography, materials for laser active substances in dye lasers which oscillate in the blue-color- or violaceous-color region, and for dying clothes in combination with one or more other organic dye compounds which suit to uses to substantially absorb a visible light. If necessary, in combination with one or more other light absorbents capable of absorbing light in ultraviolet, visible and/or infrared regions, the light absorbents can be used in clothes in general and others including building/bedding/decorating products such as a drape, lace, casement, print, casement cloth, roll screen, shutter, shop curtain, blanket, thick bedquilt including comforter, peripheral material for the thick bedquilt, cover for the thick bedquilt, cotton for the thick bedquilt, bed sheet, cushion, pillow, pillow cover, cushion, mat, carpet, sleeping bag, tent, interior finish for car, and window glasses including car window glass; sanitary and health goods such as a paper diaper, diaper-covers, eyeglasses, monocle, and lorgnette; internal base sheets, linings, and materials for shoes; wrappers; materials for umbrellas; parasols; stuffed toys; lighting devices; filters, panels and screens for information displaying devices such as televisions and personal computers which use cathode-ray tubes, liquid crystal displays, electroluminescent displays, and plasma displays; sunglasses; sunroofs; sun visors; pet bottles; refrigerators; vinyl houses; lawns; optical fibers; prepaid cards; and windows of ovens including electric ovens. When used as wrapping materials, injection materials, and vessels for the above products, the light absorbents of the present invention prevent living bodies and products from problems and discomforts

induced by environmental lights such as natural- and artificial-lights or even minimize the above them. Furthermore, they can advantageously regulate the color, tint, and appearance and control the light reflected by or passed through the products

5 to a desirable color balance.

The following examples describe the preferred embodiments of the present invention:

Example 1

Styryl dye

10 Twenty milliliters(ml) of acetic anhydride and 0.6 ml of triethylamine were placed in a reaction vessel, and mixed with 4 g of 2,3,4-trimethylthiazolium=iodide and 2 g of 4-cyanobenzaldehyde. The resulting mixture was reacted at 80 °C for one hour under stirring conditions, and then was cooled down. The

15 crystals formed were collected, washed by ethanol, dissolved in a solution of methanol and chloroform under heating conditions, and then filtrated. The obtained filtrate was distilled to remove chloroform and cooled down to obtain 1.1 g of a yellow crystal of the styryl dye represented by Chemical Formula 9. When measured

20 in a conventional manner, the melting point of the crystal was 261-263 °C.

The styryl dye of this Example with remarkable optical properties has various uses in many fields including those of optical recording media as a light absorbent.

25 Example 2

Styryl dye

Forty milliliters of ethanol and 2 ml of triethylamine were

placed in a reaction vessel, and mixed with 6 g of 1,2,3,3-tetramethyl-3H-indolenium=tosylate and 3 g of 3-nitrobenzaldehyde. The resulting mixture was reacted at 80 °C for one hour under stirring conditions, and then was cooled down. The crystals formed were collected, dissolved in a solution of methanol and chloroform under heating conditions, and then filtrated. The obtained filtrate was distilled to remove chloroform and cooled down to obtain 3.4 g of an orangish yellow crystal of the styryl dye represented by Chemical Formula 40. When measured in a conventional manner, the melting point of the crystal was 233 °C.

The styryl dye of this Example with remarkable optical properties has various uses in many fields including those of optical recording media as a light absorbent.

Example 3

Styryl dye

Twenty milliliters of ethanol and a very small amount of piperidine were placed in a reaction vessel, and mixed with 5 g of 1,2-dimethylquinolium=iodide and 2.2 g of 2-formylpyridine. The resulting mixture was reacted at 80 °C for 20 min under stirring conditions, and then was cooled down. The crystals formed were collected and recrystallized in ethanol to obtain 1.3 g of an orangish yellow crystal of the styryl dye represented by Chemical Formula 34. When measured in a conventional manner, the melting point of the crystal was 199-200 °C.

The styryl dye of this Example with remarkable optical properties has various uses in many fields including those of optical recording media as a light absorbent.

Example 4

Styryl dye

Ten milliliters of dimethylformamide and 1 ml of iodomethyl
were placed in a reaction vessel, and mixed with 1 g of the styryl
dye obtained in Example 3. The resulting mixture was reacted at
100 °C for one hour under stirring conditions, and then was cooled
down. The crystals formed were collected and recrystallized in
ethanol to obtain 0.8 g of an orangish red crystal of the styryl
dye represented by Chemical Formula 36. When measured in a
conventional manner, the melting point of the crystal was 222-223 °C.

Example 5

Styryl dye

Twelve milliliters of ethanol and 0.24 ml of piperidine
were placed in a reaction vessel, and mixed with 3 g of 1,3-
diethyl-2-methyl-5,6-dichlorobenzimidazolium=tosylate and 1.1 g
of 4-nitrobenzaldehyde. The resulting mixture was reacted at 80 °C
for one hour under stirring conditions, and then was cooled down.
The crystals formed were collected, dissolved in a solution of
methanol and water under heating conditions, and then filtrated.
The obtained filtrate was distilled to concentrate to a half volume
of the solution and cooled down to obtain 1.7 g of a yellow crystal
of the styryl dye represented by Chemical Formula 23. When measured
in a conventional manner, the melting point of the crystal was
287-290 °C.

The styryl dye of this Example with remarkable optical
properties has various uses in many fields including those of optical
recording media as a light absorbent.

Although the production conditions and yields are varied in some degrees depending on the structures of the styryl dyes of the present invention, all the styryl dyes of the present invention including the compounds represented by Chemical Formulae 1 to 60 can be produced in a satisfactory yield by the methods in Examples 1 to 5 or in accordance therewith.

Example 6

Optical property of styryl dye

Example 6-1

Light absorption characteristics of styryl dye

The styryl dyes of the present invention represented by Chemical Formulae 8, 9, 19, 23, 34, 36, 38, 40, and 44 were measured for visible absorption spectra when dissolved in methanol and formed into thin layers over glasses. In parallel, the conventional related compound represented by Chemical Formula 61 was measured for visible absorption spectrum when formed in a solution and in a thin layer similarly as above. The results were tabulated in Table 1 and FIGs. 1 to 3 show visible absorption spectra of the styryl dye of the present invention represented by Chemical Formulae 8, 36, and 61 when formed in a solution and in a thin layer, respectively.

Chemical Formula 61:

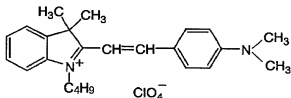


Table 1

Styryl dye	Maximum absorption(nm)		Remarks
	Solution	Thin layer	
Chemical Formula 8	354	354	Present invention
Chemical Formula 9	350	368	Present invention
Chemical Formula 19	359	366	Present invention
Chemical Formula 23	334	346	Present invention
Chemical Formula 34	364	376	Present invention
Chemical Formula 36	346	357	Present invention
Chemical Formula 38	375	430	Present invention
Chemical Formula 40	371	383	Present invention
Chemical Formula 44	372	405	Present invention
Chemical Formula 61	550	542, 574	Control

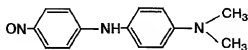
As shown in the visible absorption spectra of Fig. 3, the conventional related compound represented by Chemical Formula 61 has absorption maxima at a wavelength longer than 400 nm when formed in a solution and in a thin layer. While, as shown in the results of Figs. 1 and 2 and Table 1, most of the styryl dyes of the present invention have absorption maxima at a wavelength of 400 nm or less when formed in a solution and in a thin layer, and the absorption ends to the side of longer-wavelength area extended to around 450 nm when formed in a thin layer. These results showed that the styryl dyes of the present invention were different from conventional related compounds and that they were sensitive to a laser beam with a wavelength of 450 nm or less, particularly, a laser beam with a wavelength around 350-450 nm, and substantially absorb such a laser beam.

Example 6-2

Light-resistance improvement of styryl dye

Fifteen milligrams (mg) of either of the styryl dyes of

the present invention represented by Chemical Formulae 9, 19, 23, 34, 36, 38, 40, and 44 was added to three milliliters (ml) of TFP. To the mixture was added 2 mg of nitrosodiphenylamine represented by Chemical Formula 62 which used in the conventional optical recording media as a light-resistant improver, followed by 5-min ultrasonic energization at ambient temperature to dissolve the contents in the solvent. Thereafter, in a conventional manner, a prescribed amount of the resulting solution was dropped on either surface of a polished glass substrate, 5 cm × 5 cm, while the glass substrate was rotated at a rotation rate of 1,200 rpm for one minute to uniformly coat the solution thereupon, and sequentially blown with hot air and cold air to form a thin layer on the glass substrate. Chemical Formula 62:



The resulting glass substrate coated with either of the styryl dyes was measured for transmittance (T_0) at a wavelength of 400 nm, and then set to the position seven cm away from a 500 W xenon lamp and exposed with the light of the lamp for 25 min while sending a cold air to the substrate. Immediately after that, the resulting substrate was remeasured for transmittance (T) at a wavelength of 400 nm, and the transmittance of T and T_0 for each styryl dye was substituted for the Equation 1 to calculate the residual percentage (%) of the styryl dye. In parallel, control systems with no light-resistant improver for each styryl dye were provided.

Equation 1:

$$\text{Residual percentage of styryl dye}(\%) = \frac{100-T}{100-T_0} \times 100$$

Table 2

Styryl dye	Residual percentage of styryl dye(%)	
	With light-resistant improver	With no light-resistant improver
Chemical Formula 9	91.1	44.8
Chemical Formula 19	76.0	45.9
Chemical Formula 23	96.6	45.6
Chemical Formula 34	90.0	86.1
Chemical Formula 36	99.9	99.0
Chemical Formula 38	93.4	30.0
Chemical Formula 40	97.6	96.0
Chemical Formula 44	87.1	61.0

As shown in the results of Table 2, in the system with no light-resistant improvers, maximum 70% of the styryl dyes had changed with only 25-min exposure of light to be incapable of exerting their inherent light absorbent properties. While, when used in combination with the nitroso compound represented by Chemical Formula 62, over 76 % of the styryl dyes still remained intact without being changed by the exposure. These results indicate that light-resistant improvers are useful to suppress undesirable changes of the styryl dyes of the present invention by exposure to natural- and artificial-lights.

Example 7

Optical recording medium

Either of the styryl dyes of the present invention represented by Chemical Formulae 8, 9, 19, 23, 34, 36, 38, 40, and 44 as a light absorbent was admixed with TFP to give a concentration of 3.0%(w/w), and the mixture was further added the nitroso compound represented by Chemical Formula 62 to give a concentration of 0.35%(w/w), heated for a while, followed by ultrasonically dissolving the contents. The resulting solution was in a conventional manner filtrated by a membrane, coated in a rotatory manner over one side of an acryl disc substrate, 12 cm in diameter, to which had been transferred concaves for expressing synchronizing signals and addresses of tracks and sectors by an injection molding, to give a thickness of 200 nm. Thereafter, the substrate was spattered with silver to form a reflection layer, 100 nm in thickness, to be closely attached on the surface of the recording layer by vaporization, and the reflection layer was homogeneously coated in

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a rotatory manner with "DAICURE CLEAR SD1700", as a known ultraviolet ray hardening resin commercialized by Dainippon Ink and Chemicals, Inc., Tokyo, Japan, and irradiated to form a protection layer to be closely attached on the surface of the reflection layer, followed by obtaining nine types of optical recording media.

All of the optical recording media of this Example can be written large amounts of information in the form of documents, images, and voices at a relatively-high density by using a laser element with an oscillation wavelength of 450 nm or less.

Industrial Applicability

As described above, the present invention was made based on the creation of novel styryl dyes and the findings of their industrially usable characteristics. Since the styryl dyes of the present invention have absorption maxima at a wavelength of 400 nm or less and substantially absorb a visible light with a wavelength around 400 nm, they have diversified uses in the fields of optical recording media, photochemical polymerization, dye laser, solar batteries, lithography, dyeing, etc., which need organic compounds with such properties. Particularly, among the styryl dyes, they, which are sensitive to a laser beam with a wavelength of 450 nm or less, are advantageously useful as a material for a recording layer of high-density optical recording media such as DVD-Rs.

As compared with the conventional DVD-Rs which use polymethine dyes as a light absorbent and write information by using a laser beam with a wavelength of 635 nm or 650 nm, the optical

recording media of the present invention, which include the styryl dyes of the present invention and use a laser beam with a wavelength of 450 nm or less as a writing light, can form minute pits at a relatively-narrower track pitch and a relatively-high density.

5 Accordingly, when used the optical recording media of the present invention, they can record a vast amount of information of characters, images, and voices at a relatively-high density, resulting in greatly lowering the cost of recording information per bit and recording animations for a long time, which need much recording
10 volume, of course standing picture as a merit.

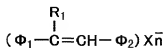
The useful styryl dyes can be easily prepared in a desirable amount by the process of the present invention which comprises a step of reacting a quaternary ammonium salt of nitrogen heterocyclic compound having an active methyl- or active methylene-group with
15 an aldehyde compound.

The present invention having such outstanding effects and functions is a significant invention that will greatly contribute to this art.

WE CLAIM:

1. A styryl dye represented by Formula 1:

Formula 1:



wherein Formula 1, Φ_1 represents a heterocycle represented by any one of Formulae 2 to 8; Φ_2 represents an optionally substituted aromatic ring or heterocycle; R_1 represents a hydrogen atom, an aliphatic hydrocarbon group, ether group, acyl group, halogen, or cyano group, and the aliphatic hydrocarbon group, ether group, and acyl group may have a substituent; X^n represents an arbitrary counter ion; and "n" is a number of X^n for keeping the balance of an electric charge in the styryl dye:

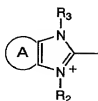
Formula 2:



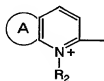
Formula 3:



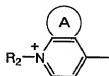
Formula 4:



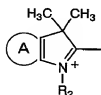
Formula 5:



5 Formula 6:



Formula 7:



Formula 8:



10

throughout Formulae 2 to 7, A represents an optionally substituted monocyclic- or polycyclic-aromatic ring or heterocycle; when A does not exist in Formulae 2 to 7, a substituent similar to that combines

for A may position to where A may exist; throughout Formulae 2 to 8, R₂ represents an optionally substituted aliphatic hydrocarbon group and R₃ represents a hydrogen or an optionally substituted aliphatic hydrocarbon group which is identical to or different from R₂.

2. The styryl dye of claim 1, wherein said ϕ_2 is a nitrogen-atom-containing heterocycle which forms an ammonium salt.

3. The styryl dye of claim 1 or 2, wherein said counter ion is an organic metal complex anion with a light-resistant improving ability.

4. The styryl dye of claim 1, 2 or 3, which has an absorption maximum at a wavelength of 400 nm or less.

5. The styryl dye of claim 1, 2, 3 or 4, which substantially absorbs a visible light with a wavelength of 450 nm or less when formed in a thin layer.

6. A light absorbent comprising the styryl dye of any one of claims 1 to 5.

7. The light absorbent of claim 6, which further contains one or more other organic dye compounds that are sensitive to a visible light.

8. The light absorbent of claim 6 or 7, which further contains one or more appropriate light-resistant improvers.

9. The light absorbent of claim 6, 7 or 8, which is sensitive to a visible light with a wavelength of 450 nm or less when formed in a thin layer.

10. An optical recording medium comprising the styryl dye of any one of claims 1 to 5.

11. The optical recording medium of claim 10, which further contains one or more other organic dye compounds that are sensitive to a visible light.

12. The optical recording medium of claim 10 or 11, which
5 further contains one or more appropriate light-resistant improvers in a recording layer.

13. The optical recording medium of claim 10, 11 or 12, which uses a laser beam with a wavelength of 450 nm or less as a writing light.

10 14. A process for producing the styryl dye of any one of claims 1 to 5, comprising a step of reacting a compound, represented by Formula 9 having ϕ_1 and R_1 corresponding to Formula 1, with a compound represented by Formula 10 having ϕ_2 corresponding to Formula 1.

15 Formula 9:



Formula 10:



Abstract

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The present invention provides styryl dyes which have
absorption maxima at a wavelength of 400 nm or less and are sensitive
5 to a laser beam with a wavelength of 450 nm or less, light absorbents
and optical recording media which comprise the styryl dyes, and a
process for producing the styryl dyes which comprises a step of
reacting with an aldehyde compound a quaternary ammonium salt of
nitrogen-atom-containing heterocyclic compound having an active
10 methyl- or active methylene-group.

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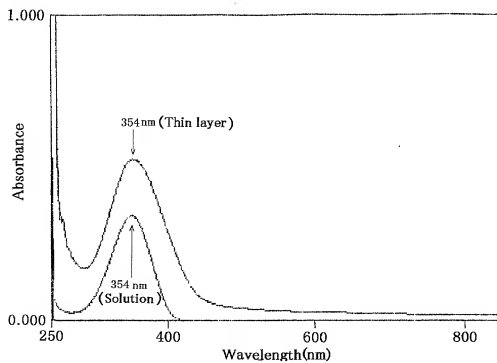


FIG. 1

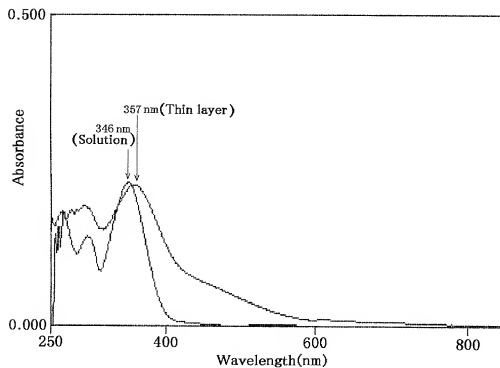


FIG. 2

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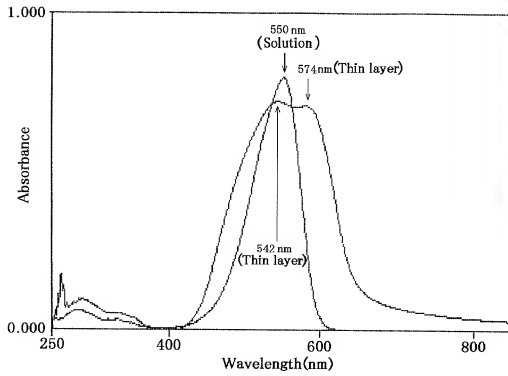


FIG. 3

Combined Declaration for Patent Application and Power of Attorney

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name; and that I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled (insert full title here) **STYRYL DYES**

the specification of which (check one)

☐ is attached hereto;

☐ was filed in the United States under 35 U.S.C. §111 on _____, as

USSN _____; or

☒ was/will be filed in the U.S. under 35 U.S.C. §371 by entry into the U.S. national stage of an intentional (PCT) application, PCT/ JP00/08298; filed 24th November 2000, entry requested on _____; national stage application received

USSN _____; §371/§102(e) date _____ (*if known),

and was amended on _____ (if applicable).

(include dates of amendments under PCT Art. 19 and 34 if PCT)

I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above; and I acknowledge the duty to disclose to the Patent and Trademark Office (PTO) all information known by me to be material to patentability as defined in 37 C.F.R. § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §§ 119, 365 of any prior foreign application(s) for patent or inventor's certificate, or prior PCT application(s) designating a country other than the U.S., listed below with the "Yes" box checked and have also identified below any such application having a filing date before that of the application on which priority is claimed:

<u>343211/1999</u>	<u>Japan</u>	<u>2nd December 1999</u>	<input checked="" type="checkbox"/> [x]	[]
(Number)	(Country)	(Day Month Year Filed)	YES	NO
_____	_____	_____	[] []	[]
(Number)	(Country)	(Day Month Year Filed)	YES	NO
_____	_____	_____	[] []	[]
(Number)	(Country)	(Day Month Year Filed)	YES	NO

I hereby claim the benefit under 35 U.S.C. § 120 of any prior U.S. non-provisional Application(s) or prior PCT application(s) designating the U.S. listed below, or under § 119(e) of any prior U.S. provisional applications listed below, and, insofar as the subject matter of each of the claims of this application is not disclosed in such U.S. or PCT application in the manner provided by the first paragraph of 35 U.S.C. §112, I acknowledge the duty to disclose to the PTO all information as defined in 37 C.F.R. §1.56(a) which occurred between the filing date of the prior application and the national filing date of this application:

(Application Serial NO.)

(Day Month Year Filed)

(Status: patented, pending, abandoned)

I hereby appoint the following attorneys, with full power of substitution, association, and revocation, to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

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The undersigned hereby authorizes the U.S. Attorneys or Agents named herein to accept and follow instructions from SUMA PATENT OFFICE as to any action to be taken in the U.S. Patent and Trademark Office regarding this application without direct communication between the U.S. Attorney or Agent and the undersigned. In the event of a change of the persons from whom instructions may be taken, the U.S. Attorneys or Agents named herein will be so notified by the undersigned.

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01-828

Page 2 of 2
Title: STYRYL DYES

Atty.Docket:

U.S. Application filed _____, Serial No. _____
PCT Application filed _____, Serial No. _____

I hereby further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. § 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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FULL NAME OF SIXTH JOINT INVENTOR	INVENTOR'S SIGNATURE	DATE
RESIDENCE	CITIZENSHIP	
POST OFFICE ADDRESS		

ALL INVENTORS MUST REVIEW APPLICATION AND DECLARATION BEFORE SIGNING. ALL ALTERATIONS MUST BE INITIALED AND DATED BY ALL INVENTORS PRIOR TO EXECUTION. NO ALTERATIONS CAN BE MADE AFTER THE DECLARATION IS SIGNED. ALL PAGES OF DECLARATION MUST BE SEEN BY ALL INVENTORS.